The 2013 World Congress on
Advances in Nano, Biomechanics, Robotics, and Energy Research (ANBRE13)
Seoul, Korea, August 25-28, 2013

Structural and Electrical Properties of CIAGS Thin Films

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ABSTRACT

CIGS thin films are proved to be the better candidate to be used as absorber layer in thin film solar cells. Thin films of copper indium gallium aluminum selenide $Cu(In,Ga_{1-x},Al_x)Se_2$ (CIAGS) were prepared by vacuum evaporation method at room temperature. In order to achieve compound formation, the as-deposited films were heated in vacuum, and were studied at different temperatures. Addition of small amount of aluminum not only gives the provision of better electrical properties but also has reduced the compound formation temperature. CIAGS thin films are prepared on glass substrates by thermal evaporation Copper, indium, aluminum, gallium and selenium are used as source materials. Samples are annealed at different temperatures in the range 150^{o}C - 500^{o}C to study the compound formation. Bruker D8 Advance is used to study the structural properties of as-deposited and annealed samples. As-deposited results show the appearance of Indium peaks as is obvious from the layout of the deposited structure. Compound formation occurred at 250^{o}C and above. Variation in crystallite size and micro strain is also observed as a function of temperature. Electrical properties are measured using four probe method. It is observed that Al contributes positively towards the electrical properties of CIGS.

1. INTRODUCTION

Solar cells based on single crystal silicon wafers have been produced with high efficiencies but there are many disadvantages of wafer based solar cells. These are high production cost, indirect band gap of silicon and low conversion efficiency (Gu 2012). For this reason, the single crystal silicon solar cells were replaced with thin film solar cells. The advantage of using thin films is the low production cost and their potential high solar to electricity-conversion efficiency, reliability, and stability (Park 2009).

The most promising materials as absorbers in thin film solar cells are the Chalcopyrites. The band gap of $CuInSe_2$is E_g=1 eV and that of $CuGaSe_2$ E_g= 1.7 eV

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(Voorwinden 2003, Faraj 2012). Therefore, a manipulation ofthe band gap between these two extremes is possible. It allows a large amount of light to be absorbed in the material (Baji 2013).

Polycrystalline $Cu(In_{1-x}Ga_x)Se_2$ (CIGS) is used as absorber layer in thin film solar cells due to its high absorption coefficient, tunable direct band gap, and excellent optoelectronic qualities (Liang 2012, Park 2009).

There are a number of methods to prepare CIGS absorber films. These methods include evaporation techniques, such as co-evaporation, sputtering and selenization, and electro-deposition (Mitzi 2009, Faraj 2012). Until now, co-evaporation is the most successful technique for the preparation of CIGS. The cell efficiencies of about 19% have been achieved (Choi 2007, Le 2012, Ko 2013).

In the evaporation method, since the composition is dependent on as-deposited thickness of only single elements, it can be very accurately controlled (Uhl 2013). But there is one of the obvious drawbacks of this technique is the small crystallite size produced (Fernandez 2005). However, the crystallinity increases as a function of annealing temperature (Lee 2010, Gu 2012).

In this paper we report on the effect of doping aluminum to the CIGS compound. We deposited thin films of copper, indium, gallium, aluminum and selenium on silicon glass substrate using vacuum evaporation method. The elements vary in composition from x=0 to x=1 according to $Cu(In,Ga_{1-x},Al_x)Se_2$. The films were annealed for compound formation and results are obtained.

2. Experimental Details

Thin films of $Cu(In,Ga_{1-x},Al_x)Se_2$ were prepared on single crystal silicon substrate with aluminum as a back contact using vacuum evaporation technique at room temperature. The deposition sequence was Cu, In, Ga, Al and Se. Edwards 306 coating unit was used to deposit these films. Rotary pump of this coater was used to maintain the pressure up to 1×10^{-3}Torr. It then employed diffusion pump to create high vacuum of 1×10^{-6}Torr. Then the samples were heated to their melting points and were evaporated. The films were deposited with variation in concentration of aluminum from x=0.1 to x=0.9 with interval of 0.2.

Before evaporation, the samples were cleaned using acetone and in ultrasonic bath while dipped in IPA for 15 minutes. The samples were also annealed at temperature 250˚C for 15 minutes for the compound formation.

In order to study the structural properties of CIGAS films Bruker D8 Advance X-ray Diffractometer was used with CuKα radiation (λ=1.5405Å). The electrical properties of CIGAS films were studied using four probe method.

2. Results and Discussion

The thin films of CIGAS were deposited for the first time using vacuum evaporation technique. We incorporated aluminum for the first time to the CIGS to make the composition of $Cu(In,Ga_{1-x},Al_x)Se_2$. In this new work, we deposited thin films of Copper, Indium, Gallium, Aluminum and Selenium with the stacking sequence of Cu, In, Ga, Al, Se. This sequence was employed by taking into account the oxidation process which is

very common in metals i.e., Cu. A series of varying composition of Ga and Al was deposited according to the composition $Cu(In,Ga_{1-x},Al_x)Se_2$ where value of x is 0.1, 0.3, 0.5, 0.7 and 0.9. The properties of CIGAS material lies in between those of CIAS and CIGS. There is no such work done on CIGAS before as there is no literature available. The series were characterized structurally, and electrically and the obtained results showed that the CIGAS thin films have the results that are in correspondence with those of CIAS and CIGS.

For the structural characterization, the XRD patterns of CIGAS thin films are shown in the Fig. 1. In order to analyze the crystal size of the deposited films, the samples were targeted by CuKα source with wavelength 1.54060Å. Annealing was performed at 250°C for 15 minutes for the compound formation. The patterns correspond to different peaks for each composition which was obtained by plotting 2θ values vs. intensity. These patterns correspond to different compositions of Ga and Al according to the composition $Cu(In,Ga_{1-x},Al_x)Se_2$ where x= 0.1, 0.3, 0.5, 0.7 and 0.9. The fig. shows the peaks of expected compositions. The peaks correspond to different compositions of CIAS and CIGS. The main diffraction peaks of all compositions based on the XRD database for CIAS (JCPD # 24-344), the XRD peaks can be assigned to the (200) phase of crystal structure of CIAS. The other minor peaks can also be assigned to different phases of CIGS (JCPD # 40-1488). Similarly there are many other minor peaks belonging to the CIAS and CIGS.

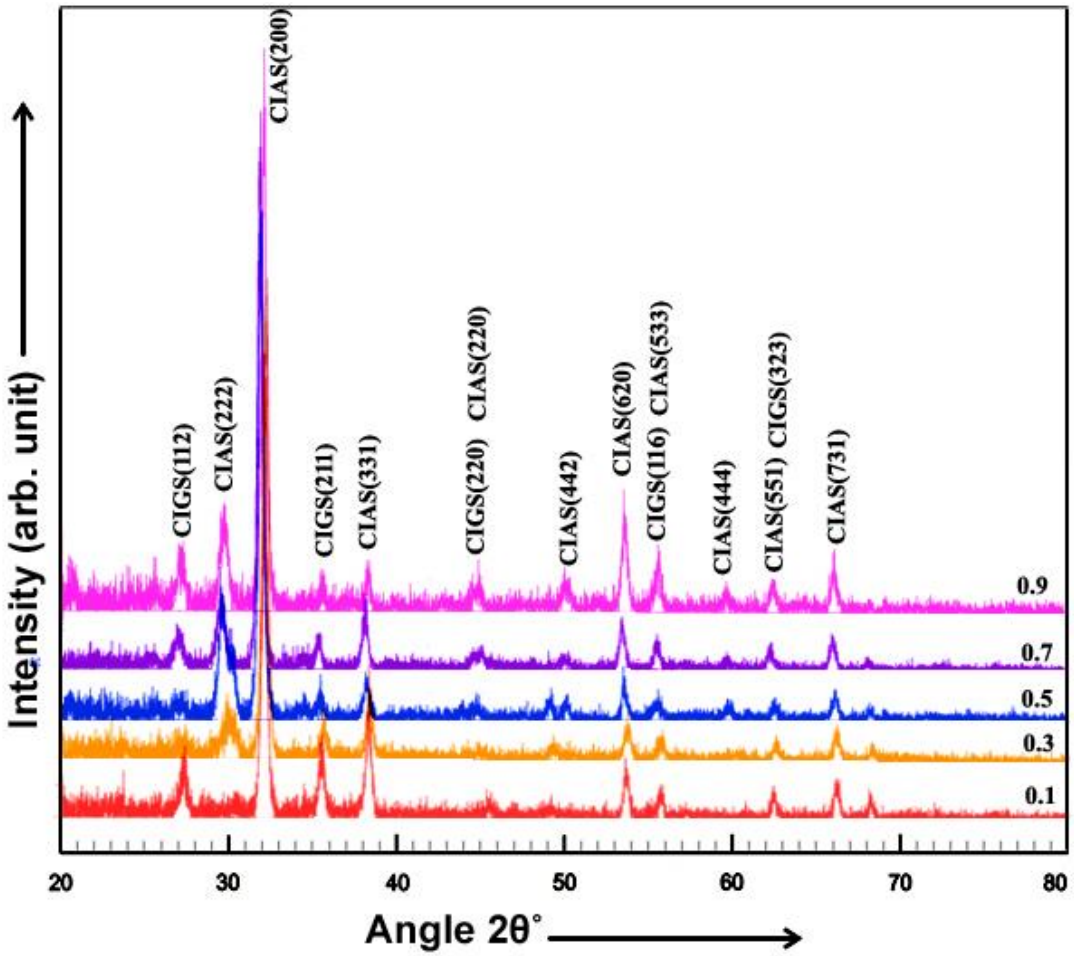

Fig. 1 XRD patterns of CIGAS films

The crystallite size of all peaks of CIGAS films was determined using Scherer equation given in Eq. (1)

$$t = \frac{0.9\lambda}{B\cos\theta} \tag{1}$$

Where B is the full width at half maximum (FWHM) of the peak, λ is the wavelength of the X-ray (1.5406 Å) and θ is the peak position. The main diffraction peaks belonging to x= 0.1, 0.3, 0.5, 0.7 and 0.9 were observed at an angle of 2θ=32.04°.

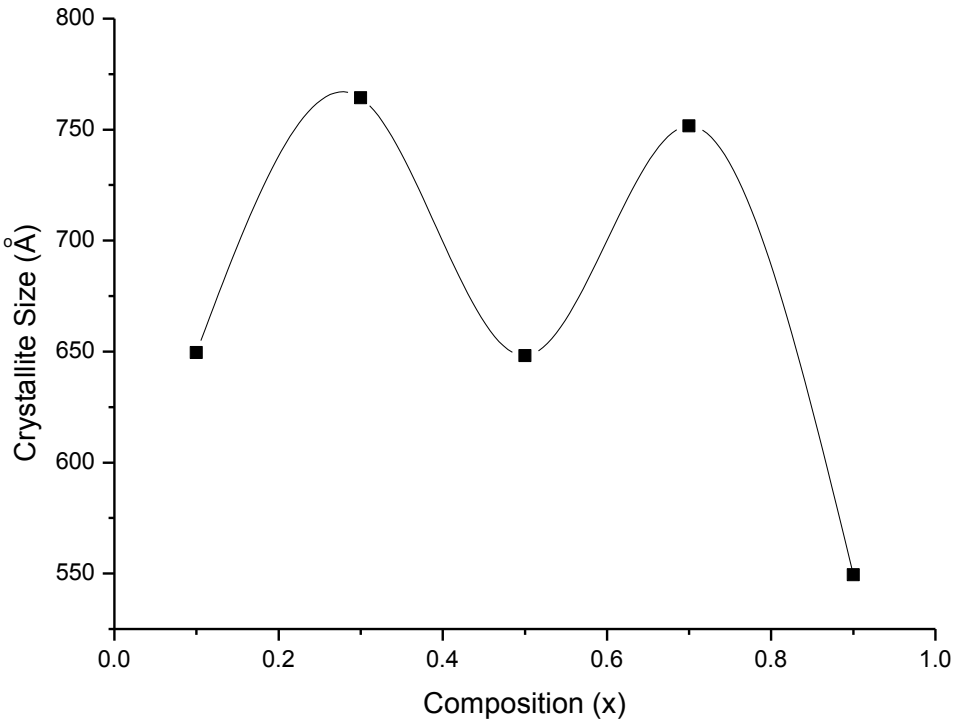

Fig. 2 Crystallite size as a function of sol concentration

The crystalline sizes for as-deposited CIGAS films was 413.45 Å for x= 0.1, 0.3, 0.5, 0.7, 0.9. The Fig. 2 corresponds to a plot between CIGAS composition and crystallite sizes. The trend shows that the crystallite size increases and then decreases forming sinusoidal curve and then decreases in the end at x=0.9.

The fig. shows a plot between intensity and different CIGAS compositions. With the increasing composition, the intensity of main peaks increases and at x=0.9, it decreases. It indicates that as the concentration of aluminum is increased the crystallinity of the films increases till concentration of x=0.7. Further increasing the concentration decreases the crystallinity of the films.

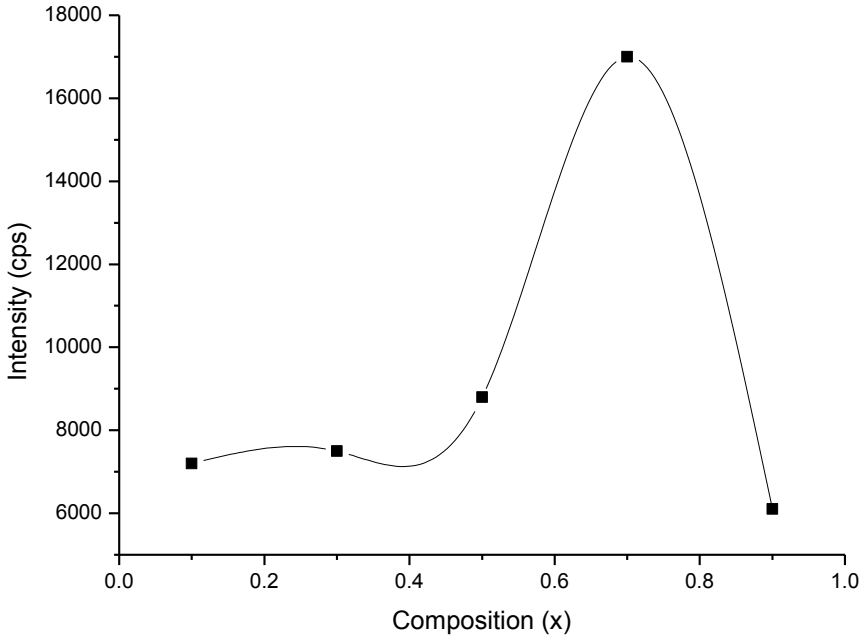

Fig. 3 Intensity of (200) plane as a function of aluminum concentration

Fig. 4 show current voltage curve for CIGAS thin films. It can be seen that electrical properties are enhanced as concentration of aluminum increases.

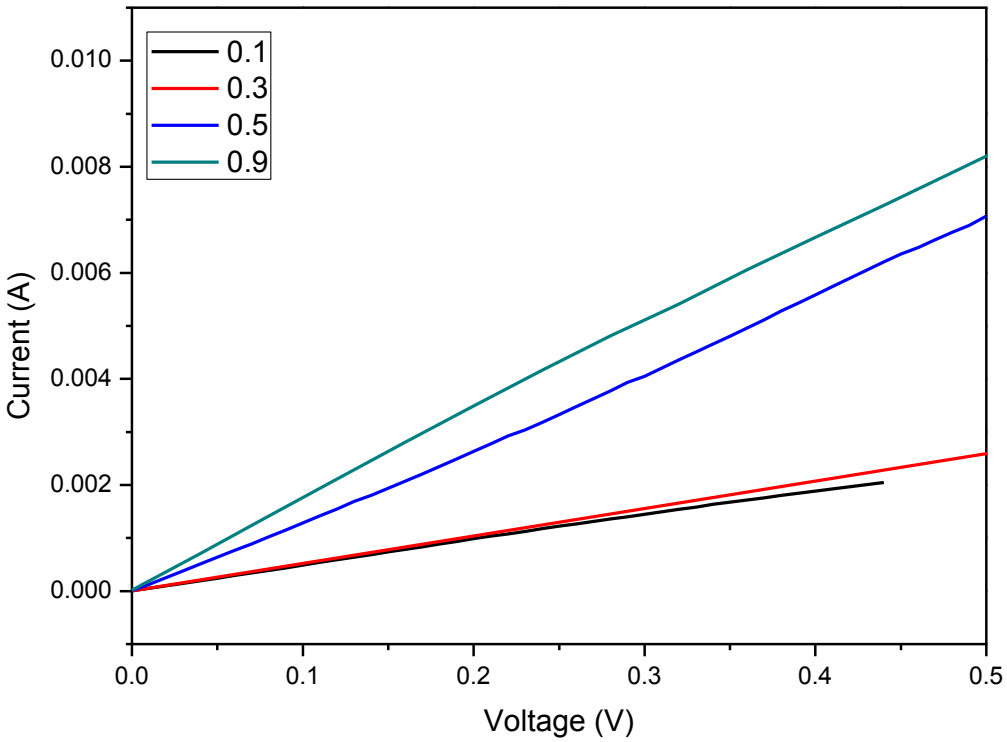

Fig. 4 Current voltage characteristic for CIGAS thin films

3. CONCLUSIONS

$Cu(In,Ga_{1-x}Al_x)Se_2$ films are prepared by sequential elemental layer technique followed by heat treatment at 250˚C. XRD patterns clearly indicate the complete compound formation. Crystallinity of the films increases till a concentration of x=0.7 where as further decreasing the aluminum concentration causes reduction in crystallinity. Current voltage curves indicate that electrical properties are enhanced with increase in aluminum concentration.

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