

STRUCTURAL, MORPHOLOGY AND ELECTRICAL STUDIES OF Cu-In-Se (CIS) THIN FILMS BY RF MAGNETRON SPUTTERING MACHINE FOR SOLAR CELLS APPLICATIONS

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ABSTRACT

CuInSe₂ (CIS) thin films can be described as one of the best candidate as absorber material for solar cell application. The CIS thin films have been prepared by radio frequency (RF) magnetron sputtering from compound ternary fan-shaped targets on low-temperature substrates with pure argon gas as the atmosphere. The stoichiometry of the ternary compound semiconductor quantum dots can easily be controlled by the ratios of the ternary elements and sputtering parameters. The CIS thin films were deposited onto FTO glass substrate in 40 min of deposition time. The annealed process of CIS thin film at 100°C in 30 min was done. The composition of CIS is obtained from a composite target consisted of copper disk of 99.99% purity, indium and selenium thin foil of 99.99% purities. The CIS nanoparticle thin films regularly shaped and distributed reasonably uniform in size on substrates CIS samples have been characterized by using Scanning Electron Microscopy (SEM) and X-ray Diffraction (XRD). The electrical properties of the samples were measured by a standard collinear four point probe technique.

INTRODUCTION

CuInSe₂ (CIS) materials with direct bandgap and a high absorption coefficient have received considerable attention as one of the most promising materials for second-generation solar cells [1]. In recent years, there has been a growing interest in CuInSe₂, an I-III-VI₂ chalcopyrite, as a material for thin films photovoltaic solar cells because of its high theoretical efficiency up to 24.8% [2]. The band gap of CuInSe₂ is about 1.04 eV, which is in good agreement with the solar spectrum. Solar cells made with CuInSe₂ have been obtained recently with an efficiency of about 15.4% [3]. They exhibit high long-term stability and can keep a relatively stable performance. These materials have also remarkably stable electrical properties over a wide range of stoichiometry.

Further success of CIS material-based solar cells use will strongly depend on the production cost of the CIS layer, since the production of this layer carries a significant part in the overall module cost. The relative high cost of the CIS layer production is

connected with the expensive vacuum technologies used for deposition layers. In order to reduce process complexity and costs of solar cells and to achieve further commercial success of CIS material based photovoltaics, as a prerequisite, the ability to produce a CIS layer on a large scale and at a low cost has to be provided. This report highlights the result of the characterization of CIS deposition layer by using RF magnetron sputtering method. RF magnetron sputtering is a low-cost and easy control method for film growth, especially suitable for large-scale film deposition. RF magnetron sputtering also can be used to sputter electrically insulating materials besides metal and alloy.

EXPERIMENTAL DETAILS

The CIS films were deposited by RF magnetron sputtering system onto FTO glass substrate at 100°C for the input power of 900W. The chamber background pressure was better than at 1×10^{-5} mbar. The deposition was carried out less than 2×10^{-3} mbar argon pressure. The composition target consists of a Cu disk with small fan shape indium and selenium plates were placed onto the Cu target disk. The diameter of the target was approximately 75 mm. Compositions of the films were adjusted by changing the number of indium and selenium once target and the substrate were mounted. Table 1 show the composition of Indium (In) and Selenium (Se) that have been mounted on Copper (Cu) target. The substrate was cleaned by extensive pre-sputtering cleaning process. The FTO glass substrate was prepared with the CIS deposition time at 40 min. After the deposited ion of CIS, the annealing process was done at 100°C for 30 mins by using oxidation and diffusion furnace. Figure 1 show the schematic of RF magnetron sputtering operated when argon gas flows. The shape and surface morphology were studied with help of high-resolution scanning electron microscopy (SEM). The microstructures were determined from X-ray diffraction (XRD). The electrical properties of the samples were measured by a standard collinear four point probe technique.

Table 1: The composition of indium (In) and selenium (Se) mounted on copper (Cu) target

Parameter	Indium (In)	Selenium (Se)	Selenium (Se)
A	1	1	Cu In(1) Se(1)
B	1	2	Cu In(1) Se(2)
C	2	1	Cu In(2) Se(1)
D	2	2	Cu In(2) Se(2)
E	2	3	Cu In(2) Se(3)
F	3	2	Cu In(3) Se(2)
G	3	3	Cu In(3) Se(3)

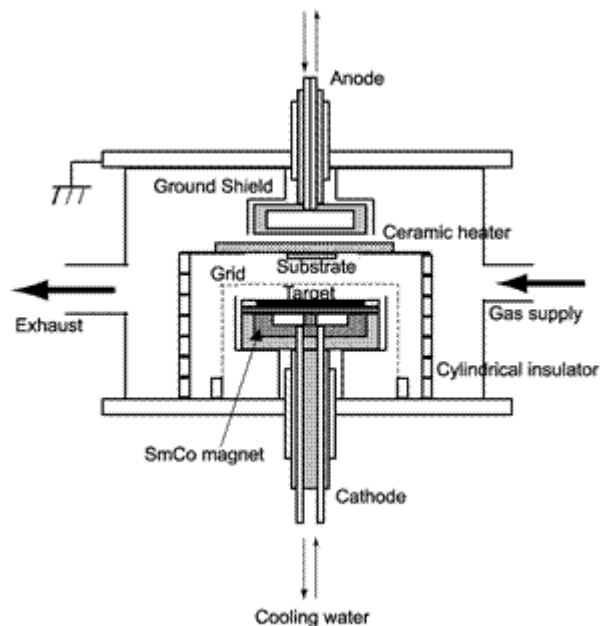
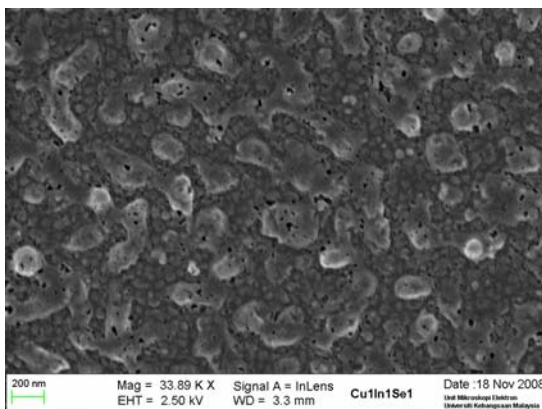


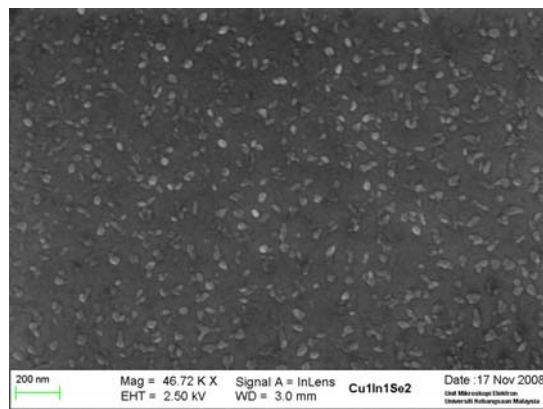
Figure 1: Schematic of RF magnetron sputtering system

RESULTS AND DISCUSSION

SEM images have been obtained for CIS thin films deposited on FTO glass substrates in order to study the surface properties. SEM brings microscopic information of the surface structure and roughness, and to determine the effect of the different indium and selenium composition on the films morphology. Figure 2 shows the SEM images of microstructure CIS thin film. SEM images of annealed CIS films showed that there is non-homogenous agglomeration and random surface texture. The grain size of CuIn(1)Se(1) is about 32.9 - 46.7 nm. A slight increase of the grain size follows increased of the composition of indium and selenium relatively. The bigger grain size of CIS thin film is around 117.2 – 150.7 nm from CuIn(3)Se(3) .



(a) CuIn(1)Se(1)



(b) CuIn(1)Se(2)

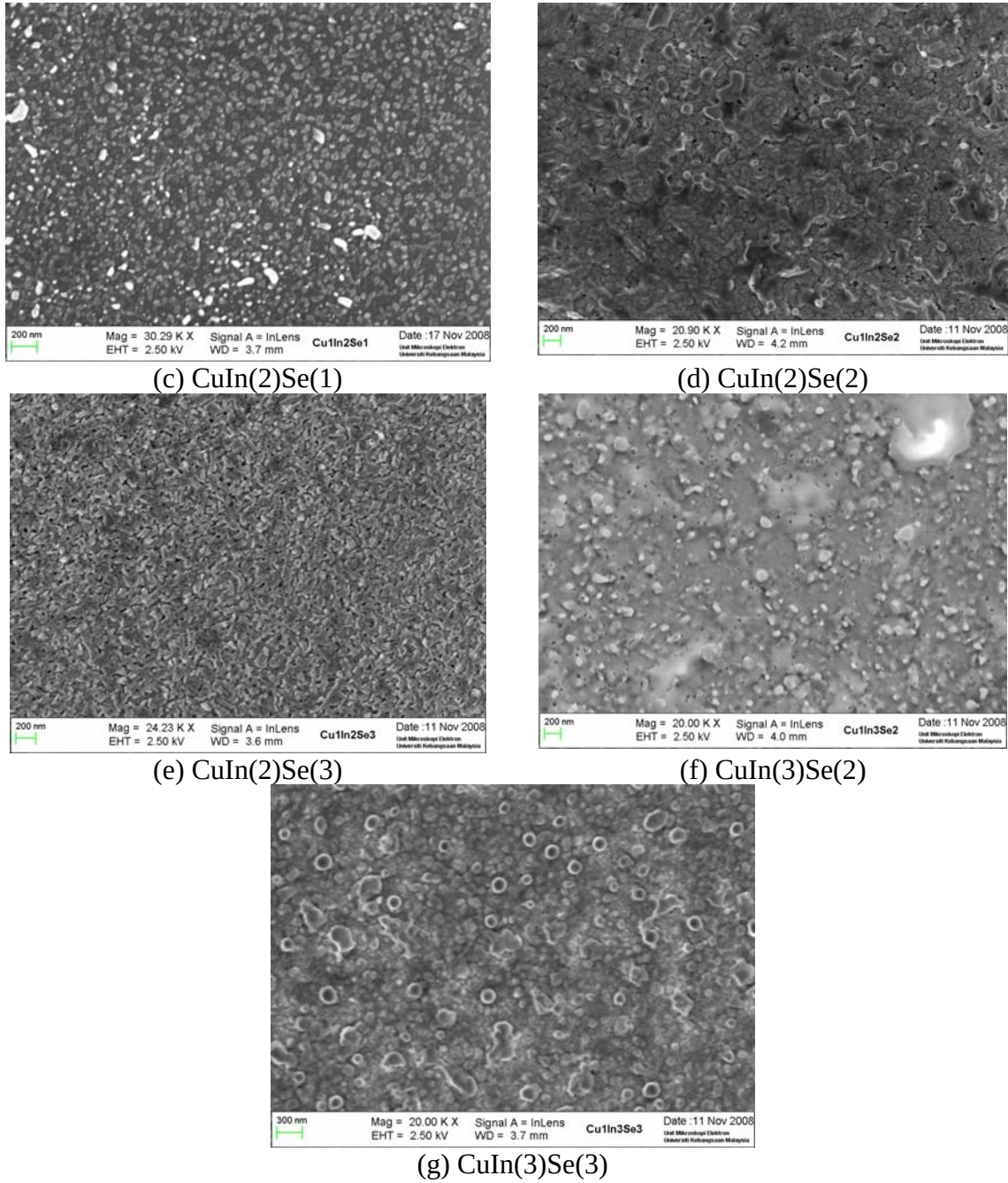


Figure 2: SEM images of nanoparticle CuInSe thin film.

The structure properties of CIS film were characterized by X-Ray Diffraction. Figure 3 below shows the XRD spectra for CIS film with different composition of indium and selenium on the FTO coated glass. After annealing the sample (100°C , 30 min, in air ambient), the ternary compound, CuInSe, and the binary compounds, CuO, FTO, or both, are formed in the deposited film.

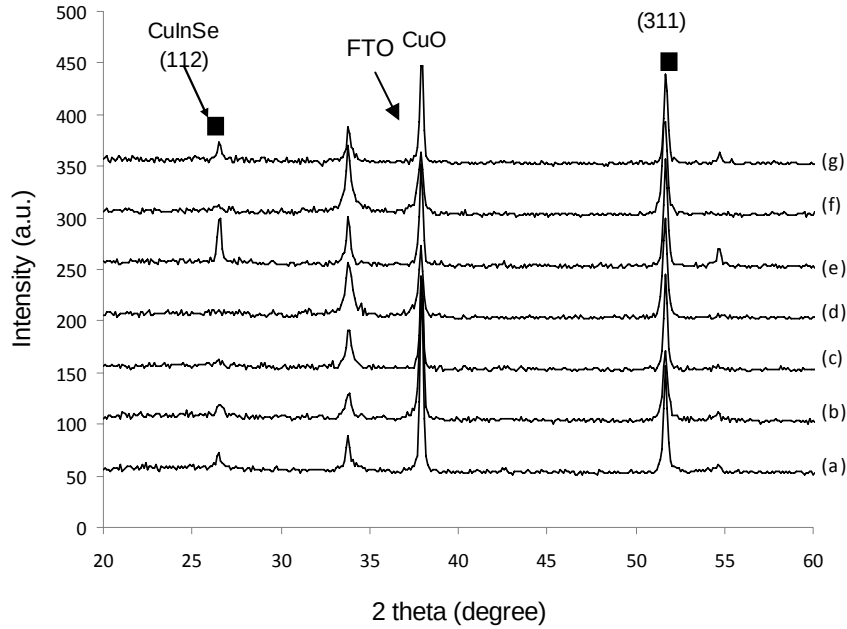


Figure 3: XRD spectra for CIS film with different composition of indium and selenium. (a) CuIn(1)Se(1) (b) CuIn(1)Se(2) (c) CuIn(2)Se(1) (d) CuIn(2)Se(2) (e) CuIn(2)Se(3) (f) CuIn(3)Se(2) (g) CuIn(3)Se(3)

The XRD spectrum for this film in Figure 3 shows several CIS peaks, some with slight shifting, indicating crystals with different selenium incorporations. XRD analysis shows the presence of CuInSe in the material. The high diffraction peaks are observed at 2 Theta of 27.1 and 53.2 and these peaks correspond to the (1 1 2) and (3 1 1) faces of cubic CuInSe, respectively. These peaks are narrowed due perhaps to differences in the different crystal compositions, with very small amounts of Indium incorporated into the structure. The peaks for FTO and CuO were also observed. CuInSe peaks are shifted to higher values due to a decrease in the lattice constant of the material.

Figure 4 shows the I-V curves for CuInSe samples measured at 100K. The sample exhibits linear I-V behaviour for forward bias condition. The slope of curve provides the resistance value which is used to calculate the surface conductivity of the samples.

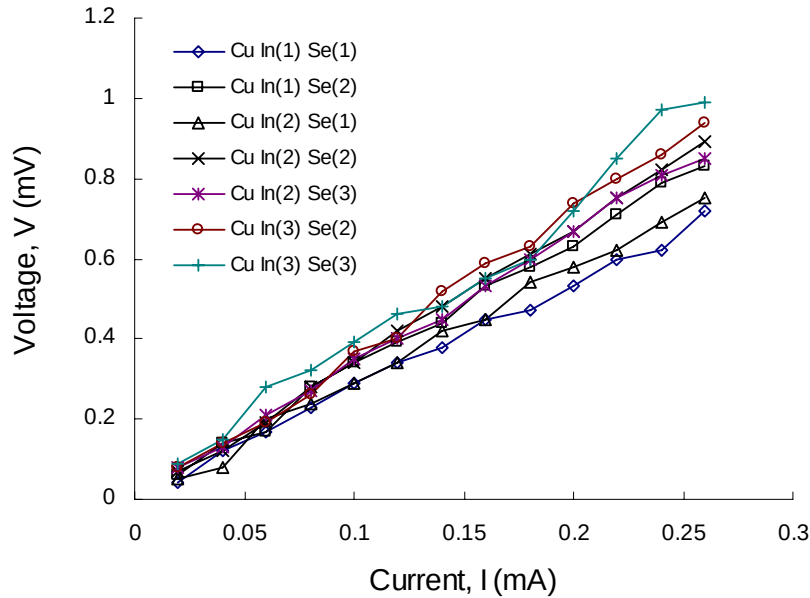


Figure 4: I-V characteristic for CuInSe samples at 300K

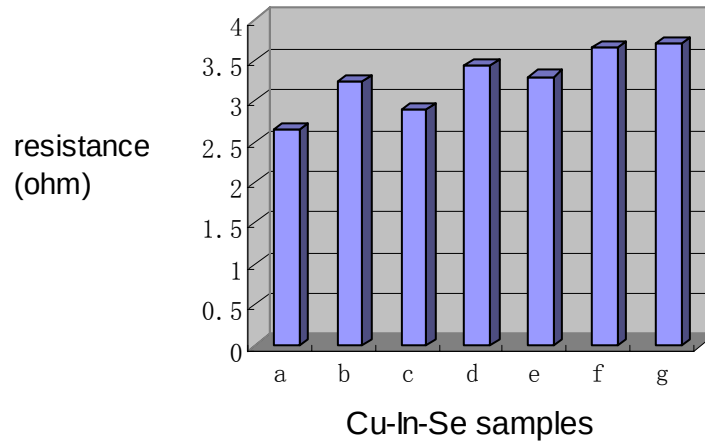


Figure 5: Histogram plot of resistance against different composition of Cu-In-Se samples. (a) CuIn(1)Se(1) (b) CuIn(1)Se(2) (c) CuIn(2)Se(1) (d) CuIn(2)Se(2) (e) CuIn(2)Se(3) (f) CuIn(3)Se(2) (g) CuIn(3)Se(3)

The samples of CuInSe thin films exhibit slightly linear I-V behaviour for forward condition. The slope of the curves provides the resistance value which is used to calculate the surface conductivity of the samples. Figure 5 shows a histogram of resistance against different composition of CuInSe samples. The resistance achieved for varied composition of CuInSe thin films are in the range of 2.5 – 3.7 ohm for CuIn(1)Se(1), CuIn(1)Se(2), CuIn(2)Se(1), CuIn(2)Se(2), CuIn(2)Se(3), CuIn(3)Se(2),

CuIn(3)Se(3) respectively. Overall, the increase in resistance follows the increase of composition in CuInSe samples. Therefore, the lower composition of CuInSe will be suitable for solar cell application as the resistance achieved is low

CONCLUSION

The CIS thin films on FTO glass substrate was prepared by using a RF magnetron sputtering machine. Annealing of the CIS thin film increases their grain size and their crystallinity. From annealed SEM images of annealed CIS films show that there is non-homogenous agglomeration and random surface texture. The grain size of CuIn(1)Se(1) is about 32.9 - 46.7 nm, meanwhile the bigger grain size is around 117.2 – 150.7 nm from CuIn(3)Se(3). Slight increase of the grain size is due to the increase of the composition of indium and selenium. XRD analysis revealed the high diffraction peaks at 2 Theta scale of 27.1 and 53.2 and these peaks correspond to the (1 1 2) and (3 1 1) faces of cubic CIS, respectively. The XRD spectrum for this film also shows several CIS peaks with slight shifts, indicating the crystallinity of this film with different selenium incorporations. The samples of CuInSe thin films exhibit slightly linear I-V behaviour for forward condition. The resistance achieved for varied composition of CuInSe thin films are in the range of 2.5 – 3.7 ohm as it increases linearly with the increase of composition of CuInSe samples. Therefore, the lower composition of CuInSe will be suitable for solar cell application as the resistance achieved is low.

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