Effect of Thickness on the Electrical Conductivity and Hall Effect Measurements of (CIGS) Films

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Abstract

The influence of different thickness (500, 1000, 1500, and 2000) nm on the electrical conductivity and Hall effect measurements have been investigated on the films of copper indium gallium selenide $CuIn_{1-x}Ga_xSe_2$ (CIGS) for x= 0.6.The films were produced using thermal evaporation technique on glass substrates at R.T from (CIGS) alloy.

The electrical conductivity (σ), the activation energies (Ea_1, Ea_2), Hall mobility and the carrier concentration are investigated and calculated as function of thickness. All films contain two types of transport mechanisms of free carriers, and increase films thickness was fond to increase the electrical conductivity whereas the activation energy (Ea) would vary with films thickness.

Hall Effect analysis results of CIGS films show all films were (p-type) and both Hall mobility and the carrier concentration increase with the increase of films thickness.

Key words: CIGS, Films, Electrical conductivity, Hall Effect, Thermal Evaporation.

Introduction

Recent environmental and energy resource have an increased interest in renewable energy sources such as photovoltaic devices. $CuIn_{1-x}Ga_xSe_2$ (CIGS) /CdS heterojunction devices are promising candidates for these applications when produced as polycrystalline materials. [1]

In yield solar cells among the various types of thin film based solar cells available in the market, CIGS film solar cells have been considered to be the most promising alternatives to crystalline silicon solar cells because of their high solar to electricity conversion efficiency up to 20.3%., reliability, radiation hardness ,and exhibit excellent stability [2,3,4].

CIGS is used as the absorber layer which is the most important layer in the PV device, reducing the thickness of absorber layer is the another approach for reducing the overall cost of the solar cell, decreasing the thickness of CIGS layer below 1 micron could lead to reduction in the production cost, with no, or only minor, loss in performance. [2]

Cu $(In,Ga)Se_2$ is a compound semiconductor exhibiting chalcopyrite crystal structure having direct band gap , large absorption coefficient ($\alpha \sim 10^5$ cm^{-1})[5,6],a moderate surface recombination velocity and radiation resistance[7].These properties give an opportunity for the fabrication of low cost, stable and high efficiency thin film solar cells[7,8]. CIGS is formed by partial substitution of In by group III element Ga. The band-gap can be changed by changing x in
$CuIn_{1-x}Ga_xSe_2$. CIGS is a self-doped (intrinsically doped) material, which means that, when the compound is formed, it automatically becomes either p- or n-type, depending upon the elemental composition present in the bulk and the surface of the film. Absorber p-type CIGS has large concentration of holes, excess electron hole pairs are generated by the light that is absorbed by the CIGS layer. [2, 6]

The primary intrinsic defects, which are also called native defects, include copper vacancies (V_{Cu}), copper-on-indium of gallium ($Cu_{In/Ga}$) antisites, which produce acceptor type defects, while indium/gallium-on-copper antisites (In/Ga_{Cu}), and selenium vacancies (V_{Se}), give rise to donor-type defects. [2]

Several methods of deposition techniques have been used to prepare CIGS films, such as co-evaporation [9,10], sputtering techniques [11], sequential evaporation and selenization [12], closed space vapor transport [13] and spray pyrolysis [14]. Even though these methods result in highly efficient CIGS solar cells, they generally require initially high capital investment as well as maintenance capital expense [2]. Many research groups show the effective performance of the CIGS based solar cell, by depositing the CIGS layer using somewhat less expensive deposition methods like flash evaporation[2], thermal evaporation[15,16], screen printing of nano particles [17], electro deposition[8],rapid thermal processor[18,19],resistive heating [5],electron- beam evaporation[20],etc.

In this research the electrical conductivity, Hall mobility and the carrier concentration of $CuIn_{1-x}Ga_xSe_2$ (CIGS) films for x= 0.6 are studied by varying films thickness from 500 nm to 2000 nm, which are prepared by thermal evaporation technique.

Experiment

$CuIn_{1-x}Ga_xSe_2$ (CIGS) films for x= 0.6 of different thickness (500, 1000, 1500, and 2000) nm were prepared by the alloy which obtained by fusing the mixture of the appropriate quantities of the elements Cu, In, Ga and Se of high purity (99.999%) in evacuated fused quartz ampoules, heated at (1273 K) for five hour.

CIGS films were prepared onto a glass slide substrate by thermal evaporation technique in a high vacuum system of ($3x10^{-6}$) torr using Edward coating unit model (E 306) from molybdenum boat. The distance from molybdenum boat to substrate was about (15 cm), the deposition rate was about (5 nm/sec) for all the films in room temperature. Al electrodes were used as contact material for making the electrical connections.

For D.C. measurement Keithly model 616 has been used to measure the variation of electric resistance (R) with temperature range (298-503) K, then calculated the resistivity (ρ) by the formula [21]:-

$$\rho=\frac{R\cdot b\cdot t}{L} \qquad (1)$$

Where t is film thickness, b is electrodes width; L is distance between two Al electrodes. The conductivity (σ) is related to the resistivity by equation [21]:-

$$\sigma_{d.c}=\frac{1}{\rho} \qquad (2)$$

Carrier concentrations and Hall mobility were calculated from resistivity and Hall voltage using equations below. Hall coefficient (R_H) is determined by measuring the Hall voltage (V_H) that generates the Hall field across the sample of thickness (t) as a function of current (I) at constant magnetic field (0.11) Tesla using Keithly model (616) according to the relation: [22]

$$R_H = V_H.t / I.B \qquad (3)$$

Where B= magnetic field

The carrier concentration is related to the Hall coefficient by equation: [22,23]

$$n_H = \pm 1 / R_H.e \qquad (4)$$

The sign of the Hall coefficient of semiconductor is determined by the sign of the charge carriers. If the conduction is due to one carrier type, we can measure the mobility according to the relation: [23]

$$\mu_H = \sigma / n_H .e = \sigma . \left| R_H \right| \qquad (5)$$

Where σ is the conductivity.

The thickness of the prepared all films was measured by using the weighing method according to the following relation[24]:

$$t= m / A .\rho \qquad (6)$$

Where: t= film thickness, m = mass of film, ρ = density of film, A= films area. Using a sensitive balance whose sensitivity of the order (10^{-4}) gm.

Results and Discussion

We can deduce from the variation in the resistivity of $CuIn_{1-x}Ga_xSe_2$ (CIGS) films for x= 0.6 which grown at R.T as a function of thicknesses (500, 1000, 1500, and 2000) nm, in Fig. (1) that the resistivity values decrease as the thickness increases due to the improvement in the films structure yields more packing density. We believe that the increase in film thickness (t) reduce dangling bonds, defects like vacancy sites, trapping centers of charge carriers and point defect cluster in the films structure, this is, perhaps, because of the decreased grain boundary scattering, therefore the resistivity of the films decrease from (4.87×10^2 Ω .cm to 1.11 Ω .cm) as the thickness increases with the improvement in the electrical conductivity σ (Fig. 2).

Figure (2) shows a plot of lnσ of CIGS films versus 10^3/T for different thickness, the activation energy of the electrical conduction can be determined. It is clear from this figure and Table 1, that the electrical conductivity increases as the film thickness increases because of the increase number of carriers available for transport for the same reasons as we mentioned before, such observations have also seen by ref. [2]. We can notice from figure (2) that all (CIGS) films have two mechanisms for electrical conductivity which means that there is two mechanism of transport of free carriers with two values of activation energy (Ea_1, Ea_2) each one predominating in a different temperature ranges. The electrical conductivity of these films is affected by the transport of free carriers in extended states beyond the mobility edge at higher temperature range (403-473) K,as well as carriers excited into the localized states at the edge of the band and hopping at other range of temperature (278-393) K, such observations have also seen by ref.[25] .

As given in Fig (3) and Table 1, the activation energy varies as the thickness increases and this is because of the improved crystal linty with the increase of the grain size.
The Hall coefficient values (R_H)were calculated for CIGS films of different thickness from equation(3) which is used to determined carrier density(n_H) and Hall mobility (μ_H) by using equation (4),(5) respectively. All CIGS films exhibit p-type conductivity, the sign of Hall coefficient for all prepared films is positive, which means that the type of conduction was p-type, i.e. holes are majority charge carriers in the conduction process, this result is in agreement with refs [1,2,6,18].In addition to that the carrier concentration into the order of $10^{17}cm^{-3}$ is in good agreement with refs[1,2,6], as well as the value of carrier mobility's in contrast with result obtained by ref.[26] which is found in the range 0.02–0.05 cm^2/Vs .All these parameters are shown in Table 2. It is clear from figures (4,5) and Table 2.that both (n_H) and (μ_H) respectively increase with the increase of thickness. This behavior can be attributed to the decrease the trapping centers of charge carriers with the increase of film thickness, this is, perhaps, because of the decreased grain boundary scattering which limits the mobility in thinner films and this is because of the improved film structure, reduce native defect centers and grain boundary defects, therefore the carrier mobility improves.

Conclusion

In research, we have used thermal evaporation method to study the effect of thickness on the electrical properties of $CuIn_{1-x}Ga_xSe_2$ (CIGS) films for x= 0.6, through measurements of conductivity and Hall Effect. From the obtained results of the present work, we conclude the following:
1-A thermal evaporation was a good method to prepare (CIGS) film at R.T from alloy.
2-The electrical conductivity and activation energies of CIGS films are seen to be dependent on the film thickness, the electrical conductivity shows as an increase of behavior with an increase of thickness.
3. The behavior of the electrical conductivity of CIGS films as a function of thickness is a result of the community between two mechanisms of transport, hopping charge transport between localized states at the edge of the band at low temperature (278-393) K and charge transport to extended state beyond the mobility gap at higher temperature (403-473) K.
4- The resistivity of these films is small; therefore these samples can be used as an absorber layer in the fabrication of solar cell.
5-Hall effect measurements confirmed that holes were predominating in the conduction process.

Both the mobility and concentration of the charge carriers increase with the increase of thickness.

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Table No. (1) : The electrical conductivity and activation energies of CIGS films at different thickness

Films thickness (nm)	$\sigma_{R.T} \times 10^{-3}$ $(\Omega.cm)^{-1}$	Ea_1 (eV)	Tem. range (K)	Ea_2 (eV)	Tem. range (K)
500	2.0519	0.013	278-393	0.147	403-473
1000	12.205	0.033	278-393	0.317	403-473
1500	92.165	0.035	278-393	0.32	403-473
2000	898.206	0.0113	278-393	0.058	403-473

Table No. (2): Values of Carrier Concentration and Carrier Mobility for CIGS Films with different thickness

Thickness (nm)	Carrier Concentration n_H (cm^{-3})	Carrier Mobility μ_H (cm^2/v. s.)
500	3.9 E+17	0.033
1000	5.4 E+17	0.142
1500	5.8 E+17	0.993
2000	1.8 E+18	3.152

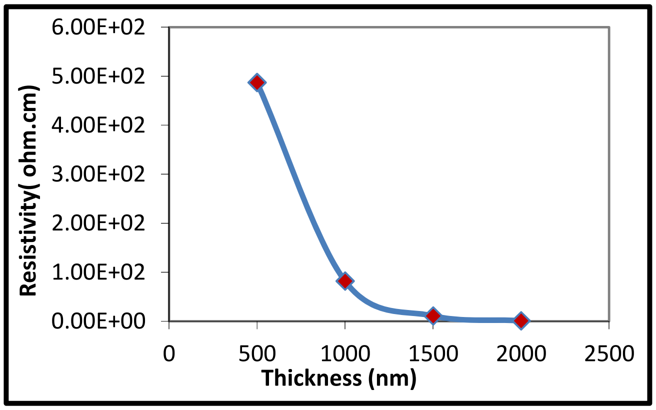

Figure No.(1): Variation resistivity of CIGS films of different thickness

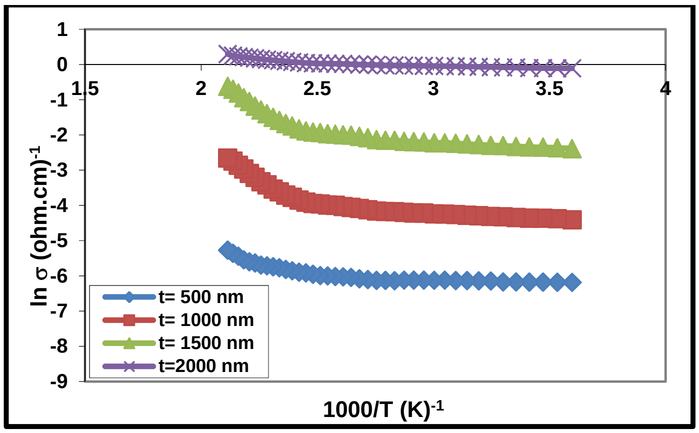

Figure No.(2): Variation lnσ versus $10^3/T$ as a function of thickness for CIGS films

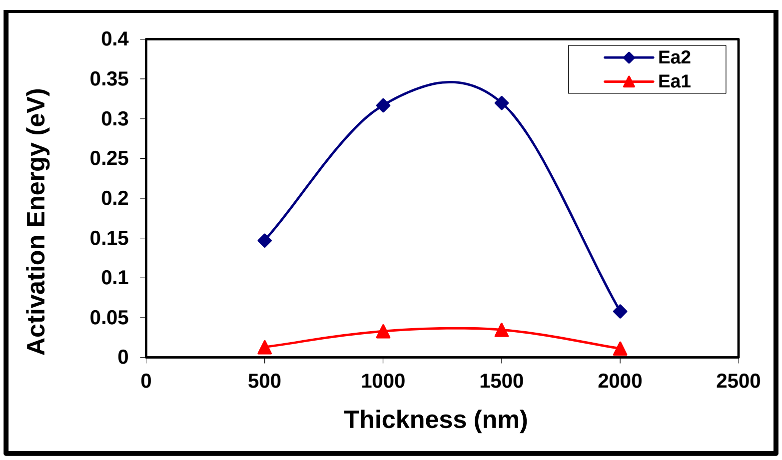

Figure No.(3): Variation activation energies as a function of thickness for (CIGS) films

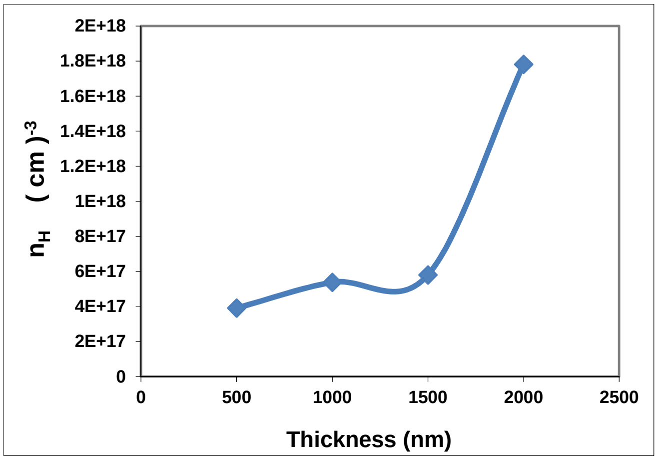

Figure No.(4): Variation of the charge Carrier's concentration For CIGS films of different thickness

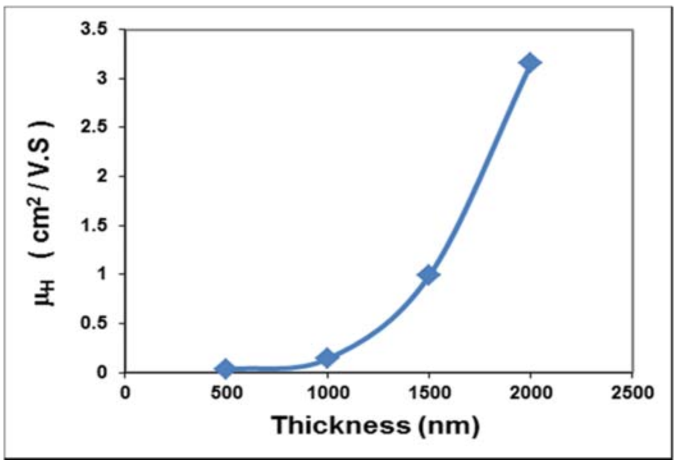

Figure No. (5): Variation Hall mobility as a function of thickness for CIGS films

تأثير السمك على التوصيلية الكهربائية وقياسات تأثير هول لأغشية (CIGS)

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الخلاصة

حسب تأثير الاسماك المختلفة nm (500, 1000, 1500, and 2000) في التوصيلية الكهربائية وقياسات تأثير هول لأغشية نحاس انديوم كاليوم سيلينيوم (CIGS) $CuIn_{1-x}Ga_xSe_2$, إذ x= 0.6. حضرت الاغشية باستعمال تقنية التبخير الحراري على ارضيات من الزجاج عند درجة حرارة الغرفة من سبيكة (CIGS).

حسبت التوصيلية الكهربائية (σ) وطاقات التنشيط (Ea_1, Ea_2) وتحركية هول وتركيز الحاملات دالة لتغير السمك. وقد أظهرت كل الاغشية آليتين للانتقال الالكتروني لحاملات الشحنة ولوحظ زيادة التوصيلية الكهربائية, بينما تنوعت طاقات التنشيط بزيادة سمك الاغشية المحضرة . وبينت نتائج قياسات تأثير هول ان جميع أغشية CIGS كانت من نوع (p-type) . كما لوحظ زيادة كل من تركيز وتحركية حاملات الشحنة بزيادة سمك الاغشية.

الكلمات المفتاحية :- CIGS ، اغشية، التوصيلية الكهربائية ، تأثير هول , التبخير الحراري.