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Optical Properties Evaluation of $CuIn_{1-x}Ga_xSe_2$ Thin Films Using OPAL2 Calculator

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Abstract: OPAL2 calculator has proved to be an effective technique for simulating optical losses in various materials for various applications. This study demonstrates the use of this software to simulate the transmittance (T) and reflectance (R) and hence evaluate the optical properties of flash evaporated $CuIn_{1-x}Ga_xSe_2$ (CIGS) thin films with a ratio x = 0.28 deposited on stainless-steel (STS) and glass substrates. The simulation results exhibit excellent accuracy of the modeled design presented in this work. The CIGS with a ratio of gallium x = 0.3 showed the best matching between simulated and experimental T and R patterns. Moreover, the simulation shows that an increase in the gallium concentration increases optical losses. Finally, the results demonstrate the application of the freeware program OPAL2 to practically simulate the optical proprieties of CIGS thin films.

Keywords: CIGS, thin films, flash evaporation, optical losses, simulation

1. Introduction

A major technological challenge in the solar energy industry is to reach the development of low-cost and efficient photovoltaic devices. A thin - film solar cell with an absorber layer based on quaternary chalcopyrite Cu (In, Ga) Se_2 compounds can meet this goal with the appropriate opto-electrical proprieties. Additionally, devices made from these materials have shown great potential with energy conversion efficiencies of the order of 23% for CIGS solar cells [1]. However, increasing the absorber band gap via higher gallium (Ga) contents x > 0.3, to minimize the effect of parasitic resistance in CIGS modules, failed because the open-circuit voltage (Voc) does not increase proportionally with the energy of the forbidden band [2]. Fundamental knowledge of the types of defects is still lacking, especially in CIGS thin films deposited on stainless-steel substrates. In this work, we will study the absorption properties of flash evaporated $CuIn_{0.72}Ga_{0.28}Se_2$ thin films, by determining the optical constants of the transmittance and reflectance

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measured by spectrophotometer UV-Vis-Nir. Also, using scanning electron microscope (SEM), the surface influence on optical losses of the elaborated samples will be investigated. Furthermore, the reflectance and transmittance will be calculated as a function of the Ga content, by simulating the optical losses in the CIGS deposited layers using an OPAL2 calculator [3]. Finally, the simulation and experimental results will be compared; a discussion is given on a possible way to accurately model the complex structures of CIGS thin films by an online freeware program.

2. Experimental Procedure

In this work, we use $CuIn_{0.72}Ga_{0.28}Se_2$ chalcopyrite thin films deposited using the flash evaporation technique onto glass and stainless-steel (STS) substrates. The source materials were prepared by the direct fusion of elements in a stoichiometric ratio and sealed in a quartz ampoule, the preparation details of the $CuIn_{0.72}Ga_{0.28}Se_2$ in ingots and thin film forms have been described previously [4]. The electrical transport properties of the obtained $CuIn_{0.72}Ga_{0.28}Se_2$ layers have been carried out at ambient temperature by Ecopia HMS-3000 measurement system based on the Hall Effect method. The optical losses were measured by a UV/Visible/Near-infrared spectrophotometer (Varian Cary 500). The thin films surface images were taken using SEM micrograph (JEOL JSM-6390LV).

3. Simulation Model

There are several simulation platforms for optical analysis like MATLAB, Ray tracing [5], OPAL 2 [3] or ANSYS based on the finite element method (FEM) technique [6]. The OPAL 2 calculator is a freeware program available on the Australian PV Lighthouse website [7]. Compared to standard modeling software where reflection, absorption and transmission (RAT) are calculated based on Fresnel's equations usually at each interaction of each ray, which considerably lengthens calculation times. OPAL2 calculator is built on the theory where all rays share a unique path reflect from the identical facets at the same angles and must consequently produce the same RAT. Thus, by regrouping the rays by their path, the resolution of the Fresnel's equations will be based not for each ray but only for each path [3]. The simulation time is drastically reduced by this simplification.

OPAL 2 is based on three [3] principal components: ray tracing, thin film optics; and equivalent-current calculations. The simulation process is presented in Fig. 1.

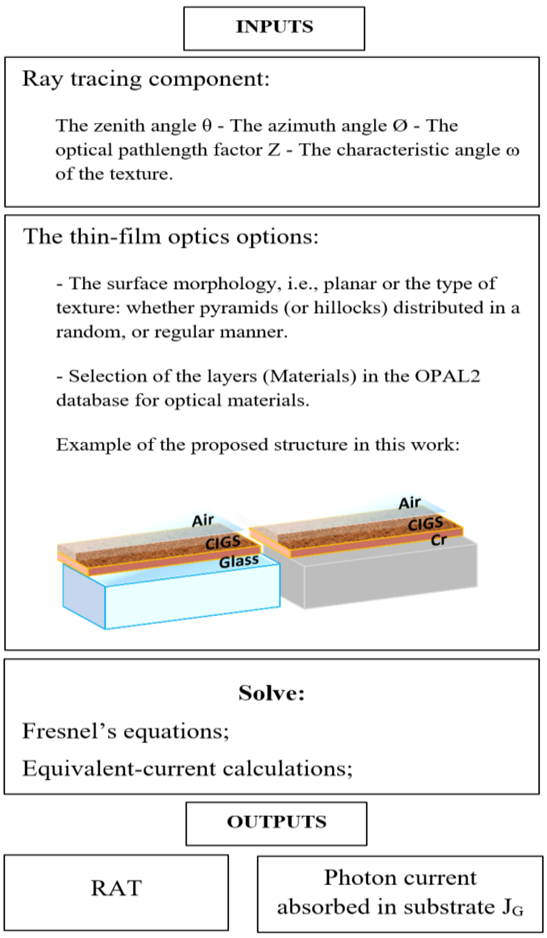


Fig. 1 - OPAL2 calculator simulation process

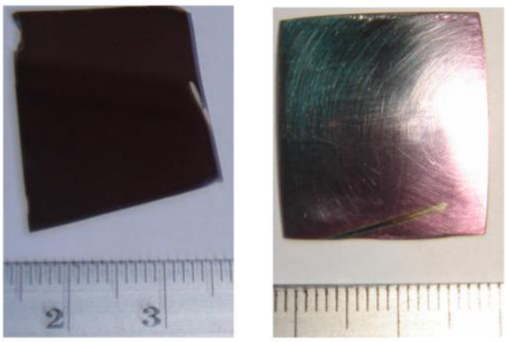
OPAL 2 calculates the transmittance, reflectance, and absorption of a thin film coating from its front surface to its substrate over a defined range of wavelengths [3,5]. The OPAL2 calculator library provides various films and substrates to model. However, the stainless-steel substrate is not proposed. To overcome this issue, we selected the element chromium (Cr) as a possible substrate replacement for the simulation since the stainless-steel used in our experiment contains a high concentration of chromium. After carefully selecting the different layers that constitute our model, we can determine the parameters of our simulation. These last are given in Table 1.

Table 1 - The simulation parameters

Ray tracing component	Surface morphology	Layer	Material	Thickness
CIGS thin films on glass substrate				
- Charact. angle, ω = / - Incident illumination: Spectrum AM1.5g Zenith angle θ = 0°	/	Superstrate	Air	/
- Light trapping model (Eq. 1): $Z = 4 + \{ln[n^2 + (1 - n^2) \times e^{-4\alpha W}]\}/\alpha W$ (1)	Planar	Film	$CuIn_{1-x}Ga_xSe_2$ [8]	140 nm
Where: α is absorption coefficient of the substrate, W is substrate thickness, and *n* is the refraction index of the film	/	Substrate	Clear soda lime [9]	/
CIGS thin films on STS substrate				
- Charact. angle, ω = 90° - Incident illumination: Spectrum AM1.5g Zenith angle θ = 0°	/	Superstrate	Air	/
- Light trapping model (Eq. 1): $Z = 4 + \{ln[n^2 + (1 - n^2) \times e^{-4\alpha W}]\}/\alpha W$ (1)	Spherical caps	Film	$CuIn_{1-x}Ga_xSe_2$ [8]	145 nm
Where: α is absorption coefficient of the substrate, W is substrate thickness, and *n* is the refraction index of the film	/	Substrate	Chromium (Cr) [10]	/

4. Results and Discussion

The flash evaporated $CuIn_{1-x}Ga_xSe_2$ thin films with x ratio = 0.28 with dimensions of approximately of 2 x 2 cm^2 are presented in Fig. 2.



(a) (b)

Fig. 2 - CIGS thin films images; (a) on a glass substrate and; (b) on stainless-steel (STS) substrate

Table 2 shows the electrical characteristics of elaborated $CuIn_{0.72}Ga_{0.28}Se_2$ films. The p-type is related to the composition with the excess of hole charge contribution. The resistivity values of CIGS layers deposited on STS substrate are lower than those of layers deposited on glass; this can be explained by the conductive effect of the STS substrate. Indeed, several studies have demonstrated that diffusion occurred between the STS substrate more precisely the steel components (Fe, Cr, Mn, etc.) and the CIGS layer [11,12]. The resistivity values obtained from CIGS layers deposited on glass substrates are in the same range as those reported by Mansour *et al.* [13].

Table 2 - Results of the resistivity of the CIGS thin films at room temperature

Substrate type	ρ (Ω cm)
Glass	2.3
STS	3.697E-3

The CIGS thin films surface images by scanning electron microscopy (SEM) (Fig. 3) allowed the defining of grain projections called spitting [14]. The formation of these spitting is due to the elevated temperature used for the evaporation of the source, especially for PVD techniques [15]. Moreover, in the flash evaporation technique [14] some parts of the source element do not turn into vapor, but remain in liquid form between the source and the substrate. They strike the surface of the substrate, forming holes or grains.

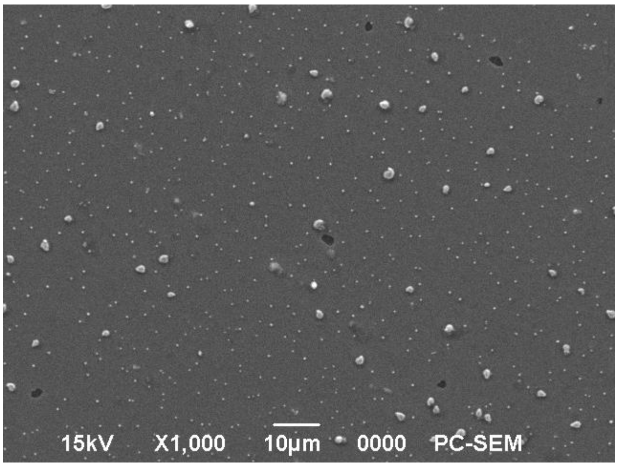


Fig. 3 - SEM micrograph of a $CuIn_{0.72}Ga_{0.28}Se_2$ thin film deposited using the flash evaporation technique

Fig. 4 presents the experimental versus the simulated transmittance (T) results of $CuIn_{1-x}Ga_xSe_2$ films deposited on soda lime glass. Additionally, the experimental and simulated T with Ga ratio x = 0.3 follow the same pattern (Fig. 4). Furthermore, an increase in T with a shift of the peak intensity from the near-infrared spectral to the visible spectral region is observed with increasing the Ga ratios. Mudryi *et al.* [16] have related this phenomenon to a decrease in the absorber surface roughness. In this work, the enhancement of T can be due to the decrease of the CIGS grain size as the Ga ratio increase, which can affect the surface properties of the absorber and consequently, the refracted light behavior [16,17].

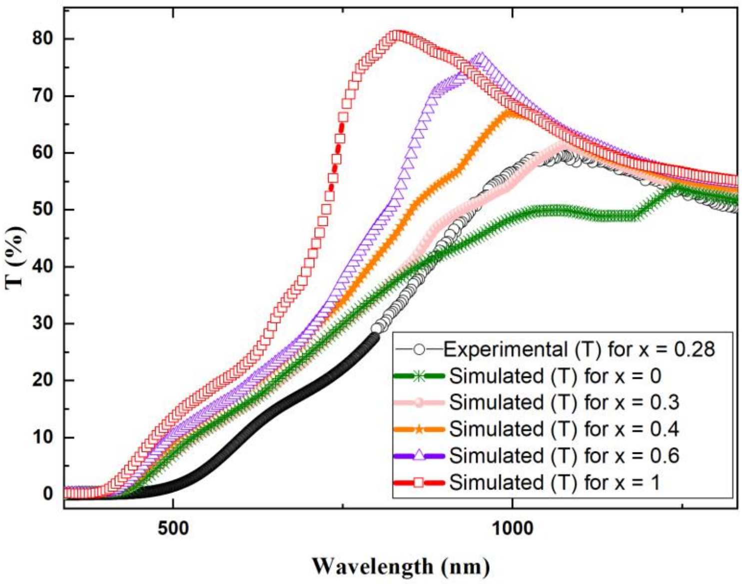


Fig. 4 - Experimental and simulated transmittance (T) spectra of $CuIn_{1-x}Ga_xSe_2$ thin films deposited on glass substrates

The experimental and the simulated reflectance spectra of CIGS thin films with different Ga ratios are demonstrated in Fig. 5. Moreover, the R simulation results with Ga ratio x between 0 and 1 follow almost the same pattern as the experimental reflectance spectra, as shown in Fig. 5, this confirming the appropriate interface selection of the simulated model. Further, the experimental maximum peak intensity is in the range of the simulated one with a Ga ratio of x = 0.3. Furthermore, the results indicate a direct relationship between the increase in the R peak intensity and Ga ratio, which confirms the reflectance influence by surface roughness and grain size [18]. Finally, this behavior is consistent with the surface image result obtained using the SEM micrograph.

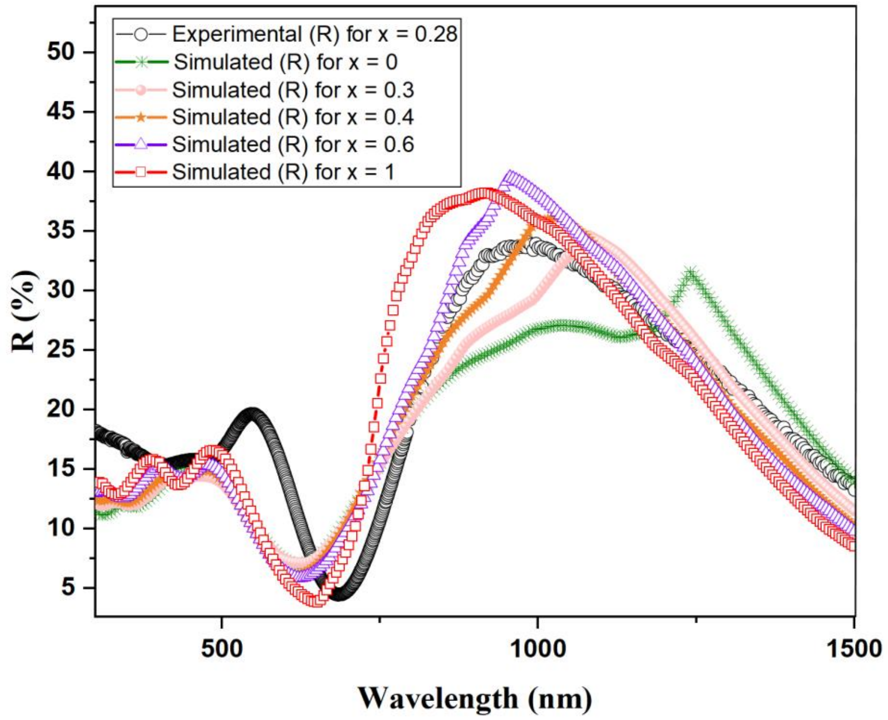


Fig. 5 - Experimental and simulated reflectance (R) spectra of $CuIn_{1-x}Ga_xSe_2$ thin films deposited on STS substrates

The band gap energies are estimated for the proportion x by extrapolating the curve $(\alpha h\nu)^2$ as a function of the photon energy (hν) (Fig. 6(a)). The values of Egg (Fig. 6(b)) are very similar to those in the literature [19,20]. Fig. 6(b) clearly shows that there is a broadening of the forbidden band with the increase in the gallium proportion [21].

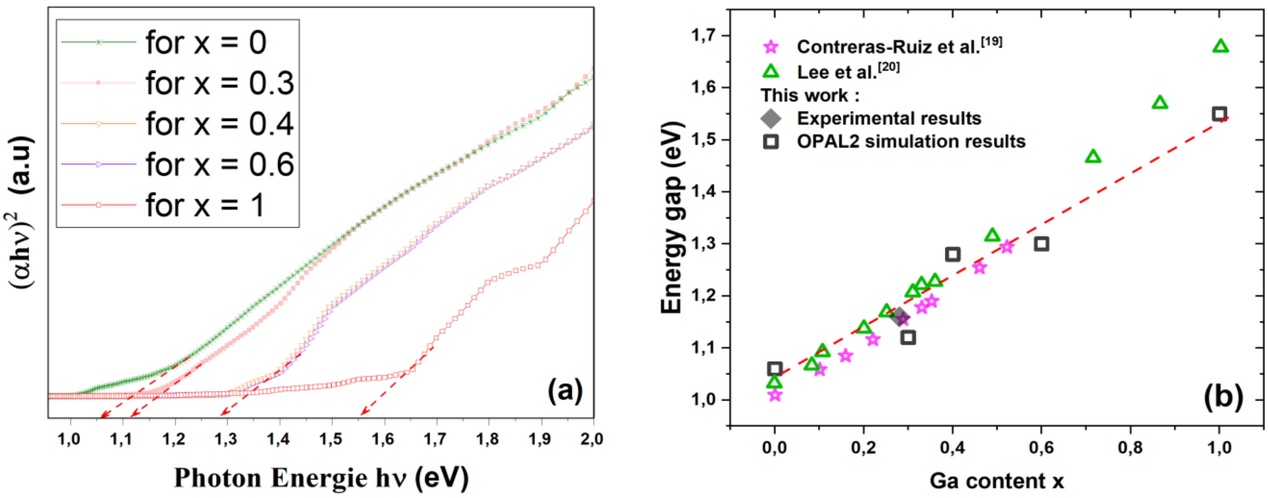


Fig. 6 – (a) Plot of $(\alpha h\nu)^2$ versus photon energy hν; (b) variation of the energy gap, as a function of composition for $CuIn_{1-x}Ga_xSe_2$ thin films

5. Conclusion

By using the OPAL2 calculator, the effects of varying the gallium (Ga) ratio on the optical properties of flash evaporated $CuIn_{1-x}Ga_xSe_2$ thin films have been investigated. A good correlation was observed between the results obtained from the simulation of T and R and those obtained using the standard spectrophotometer. Moreover, an increase in transmittance and reflectance was detected with increasing the Ga ratios. These last results were related to a reduction in the CIGS grain size, creating a smoother surface. Further, the simulated energy gap (Eg) increased with the increase in the proportion x. Finally, using the OPAL2 calculator, the analysis of the data from thin film samples is greatly simplified. The calculated and experimental optical values of flash evaporated CIGS thin films were found to be in good agreement with those reported in the literature. However, further research is necessary, especially by implementing the STS component as a substrate in the OPAL2 calculator. Furthermore, experimentally surface morphology and optical investigation of $CuIn_{1-x}Ga_xSe_2$ thin films with different Ga ratios should be carried out to check these findings.

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