Research Article

Investigation of the Optical and Electrical Properties of ITO/ZnO/CdS/CuO:Co/Ag Structure for Solar Cell

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In this work, a new structure ITO/n-ZnO/n-CdS/p-CuO:Co/Ag for solar cell was prepared on a glass/ITO substrate. The RF sputtering was used to deposit the window layer (n-ZnO) at different time periods in order to reach various thickness of this film. The n-CdS thin films were synthesized by sol-gel technique to reduce the energy bands. The buffer layer (p-CuO:Co) was sputtered at 200 W, under 30% of oxygen. Then, the electrode (Ag) with a thickness of 100 nm was deposited by thermal evaporation under a pressure of 10−5 mbar. The photovoltaic activity results obtained from this structure showed that the above method is more relevant to achieve such structure. The electrical properties of this structure were investigated using the current-voltage (I-V) and AC impedance complex measurements. The values of open circuit voltage (Voc), short-circuit current (Jsc), and fill factor (FF) are 0.46 V, 4.1 mA cm−2, and 30%, respectively. The analysis of complex impedance measurements was very useful to investigate the electrical behavior of n-ZnO/n-CdS and n-CdS/p-CuO:Co interfaces. The impedance data are presented in the Nyquist and Bode plots at different thicknesses of the n-ZnO films. An equivalent circuit was used to analyze and to fit the experimental data. The validity of these fitting results is further supported by the extrapolation and the deconvolution of both process of the diffusion and recombination processes at the n-ZnO/n-CdS and n-CdS/p-CuO:Co interfaces, respectively. Our finding could provide an efficient method for fabricating a new configuration for improving the efficiency of inorganic ZnO/CuO solar cells as well as a useful approach for the analysis of complex impedance measurements. Further works are in progress in order to better improve the conversion efficiency.

1. Introduction

During the last decades, solar cell technology has attracted the attention of many researchers. The best is not only to improve the efficiency of solar cells but also to prepare devices at a low cost. Thus, it is sought to replace the silicon and germanium by low-cost semiconductors. Cuprous oxide (CuO) is a nontoxic p-type semiconductor material with abundant soil, with optical band gap around 1.5 eV [1, 2]; cuprous oxide can act as a light absorber in heterojunction or tandem solar cells.

Copper-based oxides have promising electrical, optical, and manufacturing characteristics that establish this family of materials as well suited for semiconductor P. For this reason, CuO thin films can be obtained by different techniques such as sol-gel [3], thermal evaporation [4], electrodeposition [5], and radiofrequency (RF) sputtering technique [6].

Our previous work [1, 7] was carried out on the heterojunctions based on ZnO and CuO. However, a low conversion efficiency was obtained close to 0.01%. Therefore, the current work focused on the insertion of CdS is expected to facilitate the transport of photo-generated charge porters and the doping of the CuO by cobalt, according to the optimization achieved by our research group [8] in view to improve the conversion efficiency.

In this paper, the ITO/ZnO/CdS/Co/CuO/Ag heterojunction is prepared for the first time by the RF sputtering and spin-coating methods on indium tin oxide (ITO) substrates. Our aim is to study the effect of thickness of the ZnO film by analyzing data from optical measurements and electrochemical impedance spectroscopy (EIS).
2. Experimental Detail

ZnO window layers were deposited on glass/ITO substrates by reactive RF-sputtering in an ALCATEL SCM 451 deposition system equipped with an ALCATEL ARF 601 RF generator operating at 13.56 MHz. The ZnO thin film was then deposited at 200 W as a RF power at different times 10, 20, and 30 min. The sputtering was out in a gas mixture (argon: oxygen) with 30% of O2.

CdS thin films were synthesized on ITO/ZnO substrates in acitone using cadmium chloride (CdCl2, 0.1 M) and thiourea (CH4N2S, 0.1 M) as precursors; then, after thin films were deposited by the spin-coating method at 2200 rpm for 1 min, finally, the thin films were baked on a hot plate for 10 min to obtain the yellow color [9]. The CuO:Co thin film was deposited using an RF power of 150 W for 90 min. These parameters were obtained and used based on our previous optimization of the deposition conditions [8]. At the end, the structure was completed by depositing Ag dots on the surface of the copper oxide by thermal evaporation.

The optical properties were measured using a Shimadzu-3101-UVPC double beam spectrophotometer in the wavelength range 200–3200 nm. The electrical junction properties were investigated by current-voltage (I-V) characteristics.

The electrochemical impedance spectroscopy (EIS) was performed using a Solartron impedance analyzer model 1260 A. The measurements were carried out in the frequency range from 1 Hz to 1 MHz using the AC voltage at 50 mV. SMaRT software was used via a general-purpose interface, and Zview software was used to fit and to investigate the impedance data.

3. Results and Discussion

3.1. Analysis of the Optical Properties. The transmittance spectra of the deposited ZnO, CdS, and CuO:Co films are displayed in Figure 1. We note that the ZnO films are highly transparent in the visible range with an average transmittance value of almost 90%. Thereby, the presence of interference fringes in the transmission spectra indicates that the thin films of ZnO are homogeneous with better optical quality and the bond gap is almost constant around $E_g = 3.28$ eV (Table 1), which is in good agreement with previous works.

CuO:Co thin films are characterized by a strong absorption in the visible region, with a band Gap around 1.53 eV. While CdS thin film has an optical transmittance of 80% in the range of 600 to 800 nm, which is high enough for photovoltaic applications with $E_g = 2.40$ eV.

In addition, the alignment between the different bands (insert) is well suited for the separation of pairs ($e^-$, $e^+$), which facilitates the process of separation of charges. The band gap for CdS moves between the different components facilitating the efficient transport of electrons from the conduction band from CdS to ZnO then to ITO electrodes and transport of holes in the valence band of CuO:Co films to the Ag electrodes [10]. Therefore, this result could suggest that the 30 min cells present a better separation of the charge carriers due to the high yield of collection of the charge carriers.

3.2. Analysis of J-V Curves for a Stack ITO/ZnO/CdS/CuO:Co/Ag. The shape of the heterojunction characteristics (Figure 2) obtained under illumination is strongly linked to the thickness of the ZnO layer. The increase of deposition time showed noticeable enhancement of both open-circuit photovoltage and short-circuit photocurrent.

The parameters of the ITO/ZnO/CdS/CuO:Co/Ag solar cells such as short-circuit current JSC, open circuit potential VOC, fill factor FF (%), and efficiency η (%) at different thicknesses of zinc oxide under illumination are reported in Table 2.

The curves are in fact shifted towards increasingly larger potentials as the thickness increases. This effect may be linked to the decrease in recombination at different interfaces of the cell. ITO is a degenerate semiconductor of type n, and the electrons present in its conduction band are likely to recombine with a hole in the conduction band of CuO:Co appeared under illumination.

The impact of the ZnO layer is also clearly visible on the electrical parameters of the diodes measured under illumination. Cells with a blocking layer of thin holes (about 50 nm) have a very low current density which is the cause of the poor performance obtained. The concomitant increase in short-circuit current and open circuit potential with the thickness of i-ZnO seems to confirm the fact that this layer effectively blocks holes and passively blocks the surface of the ITO. However, an excessively thick layer of i-ZnO (40 min is not present in Figure 2) increases the series resistance significantly affecting the form factor and thus affects the performance of the solar cell. An optimal thickness is found around 150 nm ($t = 30$ min) with an efficiency of 1.1%, a short-circuit current of 4.1 mA cm$^{-2}$, an open circuit potential is 0.46 V, and a fill factor of 30%.

According to Kuo et al [11], the concentration of the supports increases with the thickness of the film, but then becomes constant when the thickness of the film is greater than 160 nm. Indeed, the 80 nm thick films have a more disordered structure and stacking defects which cause the low concentration of the carriers. These defects trap the carriers, which will reduce the concentration of free carriers. In addition, they showed that the electrical resistivity of MZO decreases first with the thickness of the film until it reaches a minimum, $1.10 \times 10^{-3}$ Ω cm, at 160 nm thick, then suddenly increases. Thus, the cells of 30 min have a better concentration of carriers with a low resistivity:

$$V_{oc} = n \left( \Phi_B + \frac{KT}{q} \ln \frac{J_{sc}}{A^*T^2} \right).$$

Hence, the increase in $V_{oc}$ as a function of the thickness of ZnO is due to the increase in the barrier $\Phi_B$ (equation (1)) due to the high concentration of the too thick layers.

3.3. Electrochemical Impedance Spectroscopy (EIS). Figure 3 show the evolutions of impedance spectra of the ITO/ZnO/CdS/CuO:Co/Ag heterojunction at different time
Figure 1: Transmission spectra of the ZnO, CdS, and CuO:Co thin films as a function of the wavelength. The inset shows the band diagrams of the isolated states of the ITO/ZnO/CdS/CuO:Co/Ag heterojunction under the equilibrium condition.

Table 1: Optical properties and preparation conditions of the ZnO, CdS, and CuO:Co thin films.

| Element   | Deposition technique | Time (min) | Gap (eV) | Thickness (nm) |
|-----------|----------------------|------------|----------|----------------|
| ZnO       | RF-sputtering        | 10         | 3.27     | 55             |
| CdS       | Spin-coating         | 2          | 2.40     | 50             |
| CuO:Co    | RF-sputtering        | 90         | 1.53     | 750            |

Figure 2: J-V curves for a stack ITO/ZnO/CdS/CuO:Co/Ag with different layer thicknesses of ZnO under illumination (600 W/m²).

periods of the deposition of ZnO in the frequency range from 1 Hz to 1 MHz. It can be seen through the impedance spectra for each time that the $Z''$ plots (Figure 3(b)) show the presence of a clear maximum at a specific frequency ($f_{\text{max}}$). This maximum shifted towards the lower frequency region when the time deposition of ZnO increased from 10 to 30 min. In addition, the magnitudes of this maximum ($Z''_{\text{max}}$) has also increased from 10 to 30 min.

The Nyquist plots (Figure 3(a)) of the impedance spectra seem to show the presence of only single semicircle. These semicircles are depressed and their diameters increased with the time deposition of ZnO. The formation of such peaks in the bode and the Nyquist plots suggests the presence of a relaxation process in the studied heterojunction, which occurs due to the characteristic frequency that separates responses between capacitive and resistive ones. It is important to note that the increase of the time deposition from 10 min to 30 min lead to increase of the thickness of ZnO layer from 55 nm to 150 nm. Therefore, it is suggested that through the impedance spectra could reflect the electrical behavior of the two n-ZnO/n-CdS and n-CdS/p-CuO:Co interfaces, respectively.

Since the evolution of the impedance spectrum showed similar behavior at different time periods, the impedance complex at $t = 30$ min was selected as typical for further analysis. As a first step, the complex impedance at $t = 30$ min was fitted with an electrical equivalent circuit including a resistance ($R_s$) in series and parallel resistance ($R_1$) with constant phase element (CPE1). This equivalent circuit is expected to describe the electrical behavior of only one relaxation process. However, it is found that the evolution of the imaginary part of impedance $Z''$ showed a large deviation between the experimental and the fitted data, as shown in Figure 4(b). In addition, the maximum observed in the experimental data do not coincide with that, for the fitted data. This deviation can also be seen in the Nyquist plot (Figure 4(a)). It is important to note that this deviation is located basically in the low-frequency region.

Therefore, a second block of parallel combination of resistance ($R_2$) with constant phase element (CPE2) was added to the previous equivalent circuit taking into account the deviation observed with only one block.

The phenomenon of depression modeled by constant phase element (CPE) is usually related to the frequency dispersion. When a CPE is placed in parallel to a resistor, a depressed semicircle is produced. The distributed capacitance is presented through constant phase element (CPE-T, P). The impedance of CPE is described as follows:

$$Z^\ast \text{(CPE)} = \frac{1}{T(j\omega)^p}.$$  \hspace{1cm} (2)

The complex impedance ($Z^\ast$) of such circuit is as follows:

$$Z^\ast (\omega) = R_s + \frac{R_1}{1 + (j\omega r_1)^{p_1}} + \frac{R_2}{1 + (j\omega r_2)^{p_2}},$$  \hspace{1cm} (3)

where $r_1$ and $r_2$ are two time constants:

$$r_1 = (R_1 T_1)^{1/p_1},$$  \hspace{1cm} (4)

$$r_2 = (R_2 T_2)^{1/p_2}.$$  \hspace{1cm} (5)

Therefore, the expression of the global impedance is the superposition of two terms similar to that representing the
Table 2: Parameters of the ITO/ZnO/CdS/CuO:Co/Ag solar cells at different thicknesses of zinc oxide under illumination.

| Sample                                    | $J_{SC}$ (mA/cm$^2$) | $V_{oc}$ (V) | FF (%) | $\eta$ (%) |
|-------------------------------------------|-----------------------|--------------|--------|------------|
| ITO/ZnO (10 min)/CdS/CuO:Co/Ag            | 0.34                  | 0.12         | 23.1   | 0.01       |
| ITO/ZnO (20 min)/CdS/CuO:Co/Ag            | 2.34                  | 0.44         | 27.10  | 0.46       |
| ITO/ZnO (30 min)/CdS/CuO:Co/Ag            | 4.1                   | 0.46         | 30     | 1.11       |

Figure 3: (a) Nyquist plot and (b) bode plots in the frequency range from 1 Hz to 1 MHz of heterojunction at different time periods of deposition of ZnO.

Figure 4: (a) Nyquist plot and (b) bode plot for the ITO/ZnO/CdS/CuO:Co/Ag heterojunction solar cell at $t = 30$ min. The insert shows the equivalent circuit for one process.
Cole–Cole relaxation in the complex permittivity formalism (equation (6)) [12]:

\[ \varepsilon^*(\omega) = \varepsilon_{\infty} + \frac{\Delta \varepsilon}{1 + (j\omega \tau)^\alpha} \]  

where \( \varepsilon_1 \) and \( \varepsilon_{\infty} \) are the low-frequency and the high-frequency dielectric constants values, respectively, \( \tau \) is the relaxation time, \( \Delta \varepsilon \) is the dielectric strength \( (\Delta \varepsilon = \varepsilon_1 - \varepsilon_{\infty}) \), and \( \alpha \) is an exponent between 0 and 1.

According to the above expression, the complex impedance, the evolution of the imaginary part \( Z^* \) as a function of the angular frequency is expected to show a maximum at a critical angular frequency \( (\omega_{\text{max}}) \). Therefore, the derivation \( ((dZ^*/d\omega) = 0) \) of the imaginary part \( Z^* \) with respect of angular frequency \( \omega \) can be used in order to find this critical angular frequency.

The above equation leads to obtain tow relationship for the angular frequency \( (\omega_{1\text{max}}) \) and \( (\omega_{2\text{max}}) \):

\[ \omega_{1\text{max}} = \frac{1}{\tau_1}, \]  
\[ \omega_{2\text{max}} = \frac{1}{\tau_2}. \]  

This circuit includes position independent distributed circuit elements relating to the transport resistance \( (R_t = R_1) \), recombination resistance \( (R_{\text{rec}} = R_2) \), and chemical capacitance (CPE: constant phase element) that are accounting for the homogeneous charge carrier transport, electron hole recombination, and capacitance per unit area due to charge accumulation associated with ITO/ZnO/CdS/CuO:Co/Ag, respectively [13]. An additional resistance \( (R_s) \) appears in the equivalent circuit attributed to the additional contribution of the series resistance. This equivalent circuit produces a good fit to the experimental data for the typical solar cell prepared at \( t = 30 \text{ min} \), as it is shown in Figure 5. Moreover, an extrapolation in the low-frequency region up to \( F = 10^{-2} \text{ Hz} \) was carried out to follow the trend of the real \( Z' \) and the imaginary part \( Z'' \) of the complex impedance. Then, a separation and/or deconvolution of the two processes were employed to further investigate the contribution and the identification of each process.

In the Nyquist plot (Figure 5(a)), the real part \( Z' \) of the impedance can be explained through three resistances: The first is found in the low-frequency region and it is corresponding to the recombination resistance \( (R_2 = R_{\text{rec}}) \). The second is attributed to transfer resistance \( (R_1 = R_t) \) in the medium frequency region, while the high frequency region is corresponding to series resistance \( (R_s) \).

These cases can be observed and checked from the Nyquists plot, where the values of \( R_1 \) and \( R_2 \) are the intercepts. The diameters of the two semicircles are corresponding to the diffusion and the recombination process at the two interfaces of the heterojunction.

The existence of the two processes is well confirmed by the deconvolution, as it is shown in Figure 5(b), on the evolution of the imaginary part \( Z'' \) as a function of frequency. Therefore, as it can be confirmed through this approach that one can see that there are two both processes of diffusion and recombination which could reflect very well the electrical behavior of the two n-ZnO/n-CdS and n-CdS/p-CuO interfaces, respectively. The diffusion and recombination processes are related to n-ZnO/n-CdS and n-CdS/p-CuO interfaces. The validity of these results is further supported by the extrapolation and deconvolution of both processes at the low and high frequency.

The equivalent circuit produces a good fit (Figure 6) to the experimental data. The fitting values of different elements in the equivalent circuit given in Figure 7 were reported in Table 3.

The diffusion time \( (\tau_1) \) and the recombination time \( \tau_2 \) corresponding to the effective electron lifetime before the recombination were calculated using the relationship extracted from equations (4) and (5) for the complex impedance.

The values of the diffusion time \( (\tau_1) \) and lifetime \( (\tau_2) \) as well as the charge collection efficiency \( (\eta_{\text{col}}) \) and the diffusion length \( (L_d) \) are plotted in histogram (Figure 8).

The obtained \( \tau_1 \) increase significantly from 11.1 ms for ITO/ZnO (10 min)/CdS/CuO:Co/Ag to 51.3 ms for ITO/ZnO (30 min)/CdS/CuO:Co/Ag by varying the thickness of ZnO. This result could suggest that the charge recombination is highly reduced in the ZnO (30 min) and the electron-hole separation [14].

The time constant of the charge diffusion is also expected to decrease with the increase of the time of the deposition of ZnO layer and/or the thickness of ZnO layer. In contrast, this later showed the inverse behavior resulting in an increase with the time (from 1.86 to 8.06 ms for 10 up to 30 min). However, this increases still very low in comparison with the time constant of the charge recombination. This could be explained probably by the formation of a very thin layer of silver oxide Ag2O3 between the active layer and contact Ag due to the oxidation of the Ag layer because the conditions for preparing the evaporator was not enough to reach \( 10^{-5} \text{ mbar} \). In these cases, the injection of electrons towards Ag can be blocked, by this barrier, and can limit the collection of charges. This defect could also explain the rasion for reaching low conversion efficiency. Similar behavior was observed by Gupta et al [13] in their study on degradation analysis of polymer/fullerene solar cells due to do formation of Al2O3, the interface between aluminium and polymer.

The charge collection efficiency \( \eta_{\text{col}} \) and the diffusion length \( L_d \) can be calculated from the \( R_{\text{rec}} \) and \( R_t \) values from [15]

\[ \eta_{\text{col}} = 1 - \frac{R_t}{R_{\text{rec}}}, \]  
\[ L_d = d \times \left( \frac{R_{\text{rec}}}{R_t} \right)^{1/2}. \]  

The charge collection efficiency \( \eta_{\text{col}} \) and the diffusion length \( L_d \) showed a very significant increase when the time of the deposition increased.
The growth in the collection yield of photo-generated carriers ($\eta_{col}$) with the time of deposition as it is reported in Table 3 could also explain the increase in JSC value obtained from J-V data as it is reported in the previous section in Table 2.

This behavior can be supported by the fact that the values of the diffusion length $L_d$ (equation (9)) are greater than the thickness of the ZnO layer (d). Thus, the majority of photo-generated carriers can reach the ITO before recombining. It appears that the increase in $V_{oc}$ is due to an increase in the density of photoelectrons in an open circuit situation, which is mainly due to the growth of $\tau_n$. Consequently, this result supports that the diffuse interface between the components of CuO:Co solar cell yield higher efficiency due to easier transport processes. The combined effect of fast electron transport, longer electron lifetime, and lower recombination rate collectively improve the $f_{sc}$ of the heterojunction [16].

Figure 5: (a) Nyquist and (b) bode plots for the ITO/ZnO/CdS/CuO:Co/Ag at $t = 30$ min. Deconvolution: low frequency (red color) and high frequency (blue color).

Figure 6: (a) Nyquist plot and (b) bode plots in the frequency range from 1 Hz to 1 MHz of heterojunction at different time periods of deposition of ZnO solid line represent the fit.

Figure 7: Equivalent circuit used to fit experimental data of the impedance spectrum at different time periods.
4. Conclusion

RF-sputtered ZnO thin films were successfully prepared using a zinc metallic target, and the effect of the thickness of ZnO window layer on the solar cell properties was investigated. The optimal time for the deposition was observed to be 30 min at a RF power 200 W with 30% of O2. Therefore, an optimal thickness was found around 150 nm which lead to obtaining an efficiency of 1.1%, short-circuit current $J_{sc} \approx 4.1 \text{ mA cm}^{-2}$, open circuit potential $V_{oc} \approx 0.46 \text{ V}$, and a fill factor of 30% (Table 2).

The electrochemical impedance spectroscopy (EIS) allowed us to investigate and to identify the diffusion and recombination processes related to n-ZnO/n-CdS and n-CdS/p-CuO interfaces. The transfers and the recombination resistance as well as the lifetime of the charge carriers were extracted from the equivalent electrical circuit. The change that occurred with the increase of window layer thickness ZnO of all these parameters reflected very well the electrical behavior of each interface.

This approach can be considered a good opportunity to prepare mainly oxide-based devices for photovoltaic applications. Indeed, these first results could make the heterojunction (ITO/ZnO/CdS/CuO:Co/Ag) as a good candidate for solar cell. It is important to state that the achieved efficiency was carried out using only a small value of illumination. Therefore, another study is in progress on the same structure using the standard illumination AM 1.5. Further studies are also expected to find the optimal absorber layer thickness in order to reach the best conversion efficiency as well as the addition of doped Al-ZnO for reducing further the series resistance.

### Data Availability

The characterization data and experimental results used to support the findings of this study are included within the article.

### Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

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