Numerical analysis and optimization of Cu$_2$O/TiO$_2$, CuO/TiO$_2$, heterojunction solar cells using SCAPS

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Abstract. In the present work, the Cu$_2$O/TiO$_2$ and CuO/TiO$_2$ heterojunction solar cells have been analyzed by the help of Solar Cell Capacitance Simulator (SCAPS). The effects of various layer parameters like thickness and defect density on the cell performance have been studied in details. Numerical analysis showed how the absorber (CuO, Cu$_2$O) and buffer (TiO$_2$) layers thickness influence the short-circuit current density (Jsc) and efficiency (η) of solar cells. Optimized solar cell structures of Cu$_2$O/TiO$_2$ and CuO/TiO$_2$ showed a potential efficiency of ~9 and ~23%, respectively, under the AM1.5G spectrum. Additionally, external quantum efficiency (EQE) curves of the CuO/TiO$_2$ and Cu$_2$O/TiO$_2$ solar cells for various layers thickness of TiO$_2$ were calculated and the optical band gap ($E_g$) for CuO and Cu$_2$O was obtained. Finally, we examined the effects of defect density on the photovoltaic parameters.

1. Introduction
Semiconducting metal oxides have been a focus of fundamental research recently [1–4]. Cu$_2$O/TiO$_2$, CuO/TiO$_2$ p–n junction have emerged as promising materials for optoelectronics and these materials are the possible solution for cheap and competitive solar cells construction. In the literature there are many reports on solar cells based on copper oxide as active layers. The conversion efficiency is the most important property in the PV domain [5,6]. Although the theoretical limit of the energy conversion efficiency of a Cu$_2$O solar cell is ~ 20% [7], the highest efficiency obtained on substrates of Cu$_2$O is 3.83% [8]. Table 1 shows values of solar cells efficiencies published in the literature. TiO$_2$ layers were fabricated by anodic oxidation of a Ti foil [9], sputtering [10], hydrothermal synthesis [8], and sintering of a TiO$_2$ nanoparticle paste [11], whereas Cu$_2$O layers were deposited by electrodeposition and sputtering [12].

In this work, we present a numerical study of the thin film solar cells Cu$_2$O/TiO$_2$ and CuO/TiO$_2$ with Solar Cell Capacitance Program. It is used to calculate the photovoltaic parameters, with the most important: short-circuit current density (Jsc) and efficiency (η) under standard illumination (AM 1.5G, 100 mW/cm$^2$, 300K).

The purpose is to study the influence the construction of the active layers on the performance of the solar cells. Towards this goal current density (Jsc) and efficiency (η) characteristics have been calculated in different thickness layer materials. Additionally, we examined the quantum efficiency
(EQE) curves in various thickness of TiO$_2$ and the effects of defect density on the photovoltaic parameters.

Table 1. Values of efficiency and heterojunction solar cells on the basis of [13-16].

| Heterojunction solar cell | Efficiency [%] |
|---------------------------|----------------|
| Cu$_2$O/ZnO              | 0.40% [13]     |
| ZnO (ZO)/Cu$_2$O         | 3.83% [8]      |
| TiO$_2$/Cu$_2$O          | 1.25%[14]      |
| TiO$_2$/CuO              | ~0.01% [15]    |
| TiO$_2$/CuO/Cu           | 0.14%, [16]    |

2. The simulation program SCAPS

There exists several numerical simulation tools for thin film solar cells [17] e.g. AFORS-HET [18], AMPS [19], ASPIN [20], SCAPS [21]. In the present investigation, we have used SCAPS-1D simulation software to study the Cu$_2$O/TiO$_2$, CuO/TiO$_2$ solar cells. There are some examples of successful thin film solar cell optimization (Frisk et al. [22]) and Zhao et al. [23]) like by layer thickness, carrier density, defect density etc. adjustment, using AMPS-1D simulator.

SCAPS is a Windows application program, developed at the University of Gent with LabWindows/CVI of National Instruments. It has been made available to university researchers in the photovoltaic community after the second PV World Conference in Wien, 1998 [24]. SCAPS version 3.2.00 is available now. Computer simulation is a valuable tool that can provide useful information for the screening process of materials to be tested experimentally [25].

The numerical simulation program SCAPS has been designed to simulate CIGS and CdTe-based thin film solar cell devices [26]. The user can calculate results in the form of following characteristics: I-V, C-V, C-f, Q(λ), band diagrams, electric field, carrier densities, partial recombination currents. The user can set parameters of materials and an operating point: temperature, voltage, frequency and illumination [27]. Device is represented as a stack of layers, up to 7 semiconductor layers with specified properties, separate entries for interface parameters and two additional layers for front and back contacts [28]. All layers are characterized by numerical description of parameters.

3. Cu$_2$O, CuO and TiO$_2$ properties

Copper forms two well-known stable oxides: cupric oxide (CuO) and cuprous oxide (Cu$_2$O). These two materials have different physical properties, different colors, crystal structures and electrical properties [29, 30]. Both forms are a natural p-type semiconductors [31].

The first attempt to use Cu$_2$O for PV application was done by National Science Foundation and Joint Centre in 1978 [25, 32]. Cuprous oxide has low electron affinity (3.2 eV) [25, 33, 34], and very high hole mobility [25,35]. Thus, it is suggested as a potential hole transport material in heterojunction based solar cells [25,36]. A few attempts were made to fabricate n-type Cu$_2$O and homo-junction Cu$_2$O cells, but the achieved efficiency is very low so far [37].

Titanium dioxide (TiO$_2$) belongs to the transition metal oxides, and is one of the best studied materials [38] in the last decade owing to its remarkable optical and electronic properties. Titanium dioxide presents good durability and a high refractive index; therefore it is suitable for applications such as antireflection coating, multilayer optical coating and optical wave-guides [39].

Properties of the materials, used in the simulations are available in the literature. The parameters of Cu$_2$O, CuO and TiO$_2$ used in our simulations are listed down in Table 2.
Table 2. The parameters values of Cu$_2$O, CuO and TiO$_2$ at 300 K on the basis of [25, 40-54].

| Material properties          | n-TiO$_2$ | p-Cu$_2$O | p-CuO  |
|------------------------------|-----------|-----------|--------|
| Band gap [eV]                | 2.26      | 2.17      | 1.51   |
| Electron affinity [eV]       | 4.20      | 3.20      | 4.07   |
| Dielectric permittivity      | 10.00     | 7.11      | 18.10  |
| (relative)                   |           |           |        |
| CB (conduction band)         | 2.0E+17   | 2.0E+17   | 2.2E+19|
| effective density of states  |           |           |        |
| [1/cm$^3$]                   |           |           |        |
| VB (valence band)            | 6.0E+17   | 1.1E+19   | 5.5E+20|
| effective density of states  |           |           |        |
| [1/cm$^3$]                   |           |           |        |
| Electron mobility [cm$^2$/Vs]| 1.0E+2    | 2.0E+2    | 10.0E+1|
| Hole mobility [cm$^2$/Vs]    | 25.0      | 8.0E+1    | 1.0E-1 |
| Shallow uniform donor density [1/cm$^3$] | 1.0E+17 | 0 | 0 |
| Shallow uniform acceptor density [1/cm$^3$] | 0 | 1.0E+18 | 1.0E+16 |

| Advantages | good transmission in the visible and near infrared regions, good adhesion, high stability against mechanical abrasion, chemical attack, high temperatures | high absorption coefficient, material abundance, non-toxicity, and low-cost fabrication | abundant, nonhazardous source materials, it can be prepared by low cost solution methods |

4. The thickness effects on the photovoltaic parameters

Absorber layer is the most important component of the solar cell where the incident photons are absorbed and excess carriers are generated. The primary role of n-buffer layer is to form a p-n junction with the p-absorber layer. The thickness of buffer layer should be as small as possible to reduce the series resistance of the PV device [55].

In the present study, the effect of absorber and buffer layers thickness on the cell efficiency ($\eta$) and short circuit current ($J_{sc}$) are simulated by changing the thickness from 1.0 to 10.0 $\mu$m (Cu$_2$O, CuO) and from 0.1 to 0.6 $\mu$m (TiO$_2$) without introducing additional defects (Figs. 2a,b, 3a,b). The proposed layer thickness is provided by the technological conditions of the thin film manufacturing technologies. A schematic structure of Cu$_2$O/TiO$_2$ and CuO/TiO$_2$ is shown in Fig. 1.
Figure 1. Schematic of heterojunction TiO$_2$/CuO(Cu$_2$O) solar cell structure with SCAPS program.

Figure 2. Conversion efficiency and short circuit current as function of buffer layer thickness for: CuO/TiO$_2$ a) and Cu$_2$O/TiO$_2$ b).

To the CuO/TiO$_2$ structure, the cell efficiency drops with an increase in TiO$_2$ layer thickness up to 0.3 μm. With further increase in buffer layer thickness, the efficiency remains nearly constant. This corresponds to a cell efficiency of 22.78%. For Cu$_2$O/TiO$_2$ the cell efficiency varies differently in relation to TiO$_2$ layer thickness, it decreases slightly and then remains constant and again drops with an increase in buffer layer thickness. Hence, for the present study we have chosen the optimal thickness of TiO$_2$ buffer layer to be 0.3 μm for Cu$_2$O/TiO$_2$ and for CuO/TiO$_2$. This corresponds to a cell efficiency of 22.78% and 9.46% respectively.

It is important to note that the curves have a similar trend in Fig.3 a,b. The solar cell efficiency increases initially with increase in CuO, Cu$_2$O layers thickness and nearly saturates at higher values. The results are in agreement with the earlier reported work [55]. When the thickness of the CuO and Cu$_2$O layer increases, the layers can absorb more photons, resulting in increased photogenerated current. Which are mainly attributed to the enhanced absorption of incident light due to the thickness of the absorber layer [56]. This saturation in efficiency is because of increased probability of SRH (Shockley-Read-Hall) recombination (due to finite carrier diffusion length) with increase in absorber thickness. Moreover, thicker absorber layer means higher material cost and fabrication cost [55]. Therefore, for the presented investigation, the optimized thickness for CuO, Cu$_2$O absorber was stated at the level of 4.0 μm. The corresponding cells efficiencies are 9.48% and 22.65% respectively.
Figure 3. Conversion efficiency and short circuit current as function of buffer layer thickness for: CuO/TiO$_2$ a) and Cu$_2$O/TiO$_2$ b).

The variation of absorption coefficient ($\alpha$) for CuO and Cu$_2$O layers was depicted in Fig. 4. The layers absorbs most of the visible light below 500 nm for Cu$_2$O and 600 nm for CuO. In the wavelength range of 500–570 for Cu$_2$O and 600–800 nm for CuO the absorption spectra decreases slightly and approaches the band gap near 600 and 820 nm respectively.

Figure 4. Absorption coefficient for CuO and Cu$_2$O thin films.

5. Quantum efficiency QE analysis
Fig. 5 presents the external quantum efficiency (EQE) curves of the CuO/TiO$_2$ (a) and Cu$_2$O/TiO$_2$ (b) solar cells for various layers thickness of TiO$_2$ (x) in the range 0.1–0.6 μm. The defect density was stated for $5 \cdot 10^{15}$/cm$^3$ for CuO and Cu$_2$O, and $1 \cdot 10^{20}$/cm$^3$ for TiO$_2$. The obtained simulations reveal that the CuO/TiO$_2$ and Cu$_2$O/TiO$_2$ exhibit significant difference in the short wavelength region < 550 nm. With further increase in wavelength, quantum efficiency falls to 0% for Cu$_2$O/TiO$_2$ in 570 nm. For CuO/TiO$_2$, it is nearly constant to 820 nm and then drops to 0%.
Figure 5a. Quantum efficiency (QE) of the Cu$_2$O/TiO$_2$ solar cell at different layers thickness of TiO$_2$.

Figure 5b. Quantum efficiency (QE) of the CuO/TiO$_2$ solar cell at different layers thickness of TiO$_2$.

The optical band gap ($E_g$) can be calculated using the model described in Ref. [57]. The calculated values and results obtain with the earlier reported works are shown in Table 3.
Table 3. Values of band gap for CuO and Cu₂O.

|                          | Calculated values [eV] | Literature results [eV] |
|--------------------------|------------------------|-------------------------|
| CuO                      | 1.40                   | 1.51 [8, 29, 49, 53, 54] |
| Cu₂O                     | 2.38                   | 2.17 [25, 41-45]        |

6. Impact of defect density in Cu₂O/TiO₂ and CuO/TiO₂ thin film solar cell

The junction and interface layers quality are very important to cell performance, because defects can debase the quality and cause much recombination. Due to the difficulty in evaluation the recombination caused by the interface defects, two hypothetical defect layers are inserted to study the interface defects [58]. These defects could affect the structural, electronic, magnetic and optical properties [38]. In the simulation, the defects densities of n-TiO₂ and for p-CuO and p-Cu₂O were controlled in the range 1E+12 to 1E+22 cm⁻³. Fig. 6 shows efficiency (η) with different defect densities of n-TiO₂, p-Cu₂O and p-CuO layer.

Figure 6. Efficiency in Cu₂O/TiO₂, CuO/TiO₂ solar cell as a function of defects density in: p-Cu₂O, n-TiO₂ layer, p-CuO layer.

The higher defect densities bring more traps and recombination centers, and deteriorate the performance of cells [58]. We note that the defect density of p-CuO layer has the most prominent influence on the device performance.

The obtained simulations reveal that for the defect density of layers <1E+18 (n-TiO₂) and <1E+16 (p-Cu₂O) the efficiency and the short circuit current remain nearly constant. With further increase of the defects density the cell parameters drop abruptly. For p-CuO layer and efficiency (η) decrease approximately linearly from 1E+12 to 1E+20.

Therefore, it is a better choice to improve Cu₂O/TiO₂ and CuO/TiO₂ interface quality by reducing defect density. The results are in agreement with the earlier reported work [58, 59].

7. Conclusion

In this investigation, the Cu₂O/TiO₂ and CuO/TiO₂ structures were studied using the solar cell simulator, SCAPS. By the analysis of the literature a complex simulation models of both structures
were completed. The solar cells were optimized for different parameters like thickness, external quantum efficiency (EQE) of buffer layer and defect density.

On the simulation basis we have demonstrated that the variation in the thickness of the absorber layer affect strongly the conversion efficiency. The photovoltaic parameters have been calculated and it was found that optimized value of the cell thickness is 4.0 μm for p-Cu₂O and CuO layers and 0.3 μm for n-TiO₂ layer.

For the defect density, we can also say that have a significant impact on the performance of Cu₂O/TiO₂ and CuO/TiO₂ solar cells. High defect density leads to pronounced decrease in the photovoltaic parameters, especially in p-CuO layer. It confirmed a high potential of both structures optimization with efficiencies of ~9% for Cu₂O/TiO₂ and ~23% for CuO/TiO₂ structure. Acquired knowledge will be practically utilized in the technology experiments, leading to the optimization cell construction.

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