Novel Transparent Conductive Oxide Bilayer Designs for Thin-Film Silicon Solar Cells

Saitta, Federica; Balaji, Aravind; Ahluwalia, Vidur; Padmakumar, Govind; Perez-Rodriguez, Paula; Santbergen, Rudi; Smets, Arno

DOI
10.1109/PVSC57443.2024.10749482

Publication date
2024

Document Version
Final published version

Published in
2024 IEEE 52nd Photovoltaic Specialist Conference, PVSC 2024

Citation (APA)
Saitta, F., Balaji, A., Ahluwalia, V., Padmakumar, G., Perez-Rodriguez, P., Santbergen, R., & Smets, A. (2024). Novel Transparent Conductive Oxide Bilayer Designs for Thin-Film Silicon Solar Cells. In *2024 IEEE 52nd Photovoltaic Specialist Conference, PVSC 2024* (pp. 766-768). (Conference Record of the IEEE Photovoltaic Specialists Conference). IEEE. https://doi.org/10.1109/PVSC57443.2024.10749482

Novel Transparent Conductive Oxide Bilayer Designs for Thin-film Silicon Solar Cells

Federica Saitta, Aravind Balaji, Vidur Ahluwalia, Govind Padmakumar, Paula Perez-Rodriguez, Rudi Santbergen and, Arno Smets

Photovoltaic Materials and Devices (PVMD) group, Delft University of Technology, Mekelweg 4, 2628CD, Delft, The Netherlands

2024 IEEE 52nd Photovoltaic Specialist Conference (PVSC) | 978-1-6654-6426-0/24/$31.00 ©2024 IEEE | DOI: 10.1109/PVSC57443.2024.10749482

Abstract — **Our study focuses on the optimization of front contact design by exploring a novel bilayer configuration that employs transparent conductive oxides (TCOs) to enhance the efficiency of thin-film silicon solar cells. The TCOs investigated include sputtered hydrogenated indium oxide (IOH), cerium-doped indium oxide (ICO), cerium and hydrogen co-doped indium oxide (ICOH), and intrinsic zinc oxide (i-ZnO). We highlight the suitability of these TCOs in a bilayer design, first analyzing their opto-electrical properties as monolayers and subsequently in bilayer configurations. The IOH/i-ZnO bilayer architecture, in particular, demonstrates promising opto-electrical properties on both flat glass and micro-textured glass substrates. IOH/i-ZnO on flat glass substrate demonstrates remarkable mobility (143.44 cm²/Vs) and a carrier concentration in the order of 10^{19} cm^{-3}. The mean of reflectance (R) trends consistently exceeds 80%, while the mean of transmittance (T) trends falls below 20% beyond 500 nm. The interference effects within the bilayers are minimized for designs on micro-textured glass, preserving values within a desirable range. These findings represent an innovative approach to front contact design for thin-film silicon solar cells, emphasizing the potential of bilayer configurations to advance solar cell technology.**

I. Introduction

The efficiency and performance enhancing of thin-film silicon solar cells has led to interest in optimizing the design of front contacts. In this study, we investigate a novel bilayer design employing transparent conductive oxides (TCOs) as the crucial materials. Specifically, hydrogenated indium oxide (IOH), cerium doped indium oxide (ICO), and cerium and hydrogen codoped indium oxide (ICOH) are considered as conductive layers, each capable of substituting the other seamlessly. In addition, intrinsic zinc oxide (i-ZnO) is included in the design for its excellent transparency in the visible part of the solar spectrum and, inherent nanostructure features.

The motivation behind investigating TCOs in a bilayer design lies in their demonstrated capability to combine very good conductivity with high optical transparency [1]. These characteristics are crucial for both efficient light absorption and effective collection of free carriers in solar cells.

Our analysis extends to a bilayer front contact on a micro-textured glass substrate. The introduction of IOH/i-ZnO bilayer configuration on such substrate demonstrates promise, as the micro-texturing aids light scattering within the solar cell, thereby improving light management and overall performance.

This work addresses to comprehensively explore the opto-electrical performance implications of different TCOs combinations in a bilayer design: emphasizing the interchangeability of IOH, ICO, and ICOH to be used alongside transparent i-ZnO.

Furthermore, the expansion to bilayer on micro-textured glass enables us to investigate the interplay between opto-electrical properties of the front contact design and the substrate morphology. This analysis therefore drives specific enhancements in the front contact design of solar cells, contributing to notable progress in the current state of thin-film technology [2].

II. Experimental Procedure

A. Fabrication

Corning glass (10×10 cm^2) is used as the substrate for TCO film depositions. Glass substrates undergo cleaning in acetone and isopropyl alcohol ultrasonic baths, each for a duration of 10 minutes. Various TCO materials are under investigation to identify optimal candidates for the bilayer TCO design. The Indium Oxide (In_2O_3) binary compound is doped with hydrogen, cerium, and a combination of both, resulting in the formation of IOH, ICO, and ICOH respectively. Additionally, the Zinc Oxide (ZnO) is employed without any impurity-doped materials. TCO layers are deposited using the Radio Frequency (RF) magnetron sputtering technique, with adjustments made to time, power (P), pressure (p), temperature (T) and oxygen content to explore various deposition conditions. In the cases of IOH and ICOH, partial H_2O pressure (H_2O p) is introduced as an additional parameter. The set and varied sputtering settings for TCO single are summarized in the following table.

TABLE I
Summary of Sputtering Settings

TCO	T [°C]	P [W]	1%O2-99%Ar flow [sccm]	p [e-3 mbar]	H_2O p [mbar]
IOH	25	150-180	/	5.7	2.0 e-5
ICO	25	200	2-4	3.0	/
ICOH	25	60	3-5	3.0	3.0 e-6
i-ZnO	200-400	200-400	/	2.6	/

To enhance film quality, TCO samples are subjected to a post-deposition annealing (PDA) treatment in an atmospheric environment. The annealing temperature (T_a) is set at 200 °C for all samples. The saturated annealing time (t_a) is optimized at 15, 20, 30 min and 2 h 20 min for ICO, IOH, ICOH and bilayer samples respectively.

B. Characterization

Given the complex interplay between optical and electrical properties, several metrics are chosen for the analysis.

Optically, the sample is characterized by refractive index (n), extinction coefficient (k), optical bandgap (E_g) and, bulk thickness (t), determined using a spectroscopic ellipsometry (SE) system. In the SE analysis, the dielectric function of TCO samples is considered homogenous in depth and modeled by combining two oscillators: Cody-Lorentz and Drude.

For bilayers, a spectrophotometric PerkinElmer Lambda 1050 system is employed to measure transmittance (T) and reflectance (R). This alternative method is chosen due to the intricacy of SE fitting using dedicated oscillator theories on bilayers, offering a practical and effective approach to assess the optical properties of the complex structure.

Electrically, the TCO samples are characterized by conductivity (σ), which is directly linked to carrier mobility (μ), free carrier concentration (N), and elementary charge (e). Hall measurements enable the determination of resistivity (ρ) using the van der Pauw method, from which N and μ are derived through the Hall effect [3]. Instead of focusing on conductivity, parameters like carrier density and mobility are employed as device-relevant parameters to evaluate the electrical properties of TCOs.

III. Results and Discussion

The front contact architecture comprises two layers. The first deposited layer, derived from multicomponent indium oxides, serves as highly conductive layer for the lateral transport and the collection of free carriers at the front part of the solar cell. The ZnO, positioned atop the former TCO, offers transparency and facilitates the transverse transfer of free carriers.

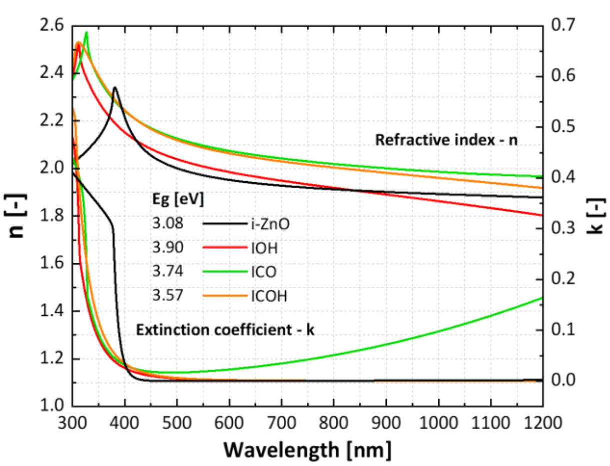

Fig. 1. Refractive index (n), extinction coefficient (k) and energy bandgap (Eg) of i-ZnO, IOH, ICO, ICOH films with optimised PDA annealing time (t_a) and T_a = 200 °C.

Enhancing conductivity is achievable through increased carrier concentration and/or mobility. However, an upper limit exists due to the constraints of dopant scattering and doping efficiency. This imposes a trade-off between N and μ, indicating that σ cannot continuously rise [4].

Furthermore, intra-band transitions within the conduction band lead to parasitic Free Carrier Absorption (FCA) in the Near-Infrared (NIR) region. In metals and semiconductors, FCA is modelled by the Drude oscillator theory, establishing a direct relationship between NIR absorption and N. The optimized trade-off yields TCO films with high mobility and limited free carrier concentration, ensuring a balanced approach from both optical and electrical perspectives.

Initially, the behaviours of the refractive index and extinction coefficient in single TCO films is examined. Figure 1 illustrates the n&k values and the energy bandgap following the optimisation of the sputtering settings and the PDA time. The trend in k can be directly correlated to the TCOs absorption coefficient through the 4πk/λ relationship. The saturated t_a for IOH and i-ZnO is 20 min reducing the FCA in the NIR region close to zero. This also holds true for the k curve of ICOH after a 30 min PDA treatment. However, no improvement is observed for ICO material with t_a longer than 15 min, indicating that the NIR region contributes to parasitic absorption in ICO. Among the investigated TCOs, IOH exhibits the lowest band-to-band absorption, resulting in an Eg of 3.9 eV.

The trends in refractive index (n) indicate a more effective refractive index grading between ICO and i-ZnO. This suggests a promising approach for reducing front reflection losses and thus, enhancing the optical performance of solar cell devices.

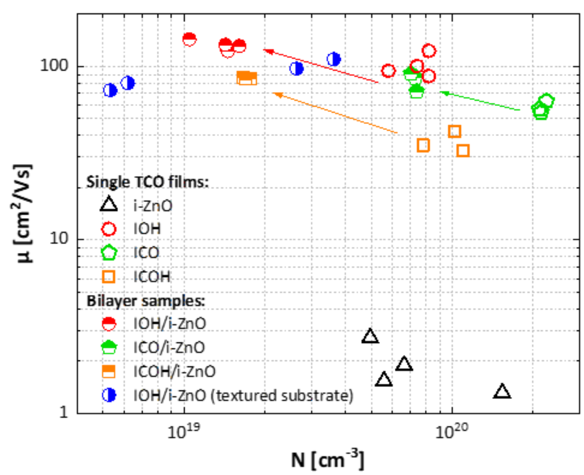

Fig. 2. Charge carrier mobility (μ) and concentration (N) of i-ZnO, IOH, ICO, ICOH monolayers (open symbols) and bilayer designs (half open symbols) on flat and micro-textured glass substrates. The presented μ and N values are shown after PDA treatment at 200 °C, with each mono- and bi-layer having its own annealing time optimized.

In Figure 2, the relationship between mobility and carrier density is depicted for monolayer TCO films using empty symbols, while the electrical properties of various bilayer designs are illustrated using half-full symbols. Each TCO material is investigated by changing the most relevant sputtering parameter and seeing how it might affect the electrical properties. The deposition time is adjusted based on the deposition rate to achieve a targeted thickness of approximately 100 nm for each TCO monolayer.

The IOH monolayers exhibit excellent mobility values, reaching up to 122.96 cm^2/Vs, with carrier concentration lower than $8.2 \cdot 10^{19}$ cm^{-3}. The ICO and ICOH samples reveal lower μ and higher N compared to IOH values. The IOH is selected as the most promising conductive candidate due to its superior electrical performance. And, it is further analyzed on flat glass and subsequently transferred onto micro-textured glass substrate. A wet etching process, involving the etching of 300 nm of Indium Tin Oxide (ITO) layer, is employed to imprint microtextures on the glass substrate.

Furthermore, the i-ZnO single layers show both low μ and N values, indicating that the scattering mechanism is mainly constrained by the grain boundaries of this material. Conversely, the low N values suggest that incorporating i-ZnO in the bilayer structure does not have an impact on the parasitic FCA.

For each bilayer structure, 1 µm of i-ZnO is sputtered under various investigated settings for IOH, ICO, and ICOH. The power and temperature for the deposition of i-ZnO are set at 200 W and 200 °C respectively. The most performing bilayer design is IOH/i-ZnO on flat glass: μ is 143.44 cm^2/Vs and N is $1.05 \cdot 10^{19}$ cm^{-3}. The values of IOH/i-ZnO structure on textured glass are comparable with the flat design and however, largely scattered. The interplay between the texture and bilayer emphasizes the importance of achieving homogeneity in texture and in TCOs deposition for efficient light scattering and optimal electrical performance of the front contact.

Figure 3 illustrates the trends in reflectance and transmittance as a function of wavelength for the investigated bilayer designs. Beyond 500 nm, the mean of T and R trends surpass 80% and fall below 20% respectively. The interference effects result from the interaction of light waves reflected within the TCO layers or between the bilayer and the substrate. Noteworthy peaks and valleys in the R and T curves of IOH/i-ZnO (flat glass substrate) and ICOH/i-ZnO highlight wavelengths with significant constructive and destructive interferences. These effects are mitigated in the ICO/i-ZnO design and further smoothed in the case of IOH/i-ZnO on a textured substrate.

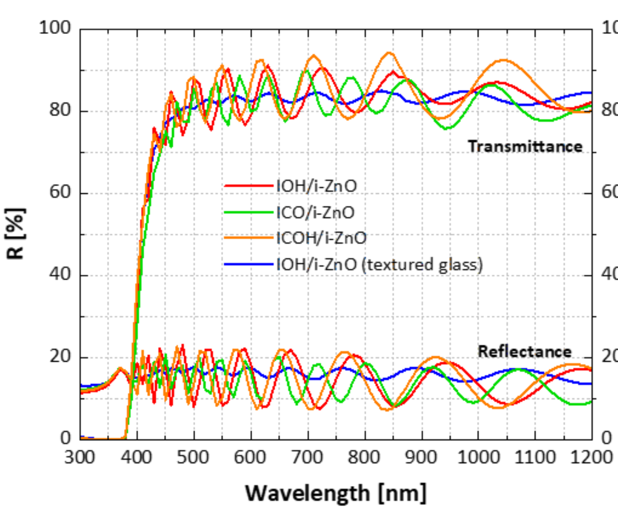

Fig. 3. Reflectance (R) and Transmittance (T) as functions of wavelength for bilayer designs: IOH/i-ZnO on both flat and textured glass, and ICO/i-ZnO, ICOH/i-ZnO on flat substrate. The thickness of IOH, ICO, ICOH layers is within the range of 100 nm, while i-ZnO is 1 µm. Trends in R and T are presented following 2 h and 20 min PDA treatment at 200 °C.

Further optimization of the bilayer design on textured glass is expected to enhance transmittance and reduce reflectance at the front contact of thin-film silicon solar cells. This refinement in light management is likely to improve the active layer absorptance (A=1–R-T), translating into better device performance.

IV. Conclusions

In conclusion, our investigation into optimizing front contact for thin-film silicon solar cells led to a novel bilayer design using transparent conductive oxides (TCOs). Hydrogenated indium oxide (IOH), cerium-doped indium oxide (ICO), and cerium and hydrogen co-doped indium oxide (ICOH) were explored, to be combined with intrinsic zinc oxide (i-ZnO). The study emphasized the interchangeability of these TCOs in a bilayer design, contributing to advancements in thin-film technology.

The bilayer architecture, particularly IOH/i-ZnO, demonstrated superior opto-electrical properties on flat glass compared to single layer architectures, and promising results on micro-textured glass were also shown. The mobility reached up to 143.44 cm^2/Vs and carrier concentration in the order of 10^{19} cm^{-3} showcasing its superior optical and electrical performance.

In addition, the reflectance (R) and transmittance (T) trends for bilayers on flat glass substrate exceeded 80% and fell below 20% respectively, beyond 500 nm. Interference effects have been smooth out preserving in the same range values of R and T for IOH/i-ZnO on micro-textured substrate.

We plan to highlight the impact of the bilayer configuration on solar cell performance during the conference. This demonstration will underscore its applicability and potential enhancements in thin-film nanocrystalline silicon (nc-Si) solar cells, contributing to the advancement in solar cell technology.

REFERENCES

[1] Bel Hadj Tahar, R., Ban, T., Ohya, Y., & Takahashi, Y., “Tin doped indium oxide thin films: Electrical properties”. Journal of Applied Physics, 83(5), 2631-2645, 1998.

[2] Tan, H., Moulin, E., Si, F. T., Schüttauf, J. W., Stuckelberger, M., Isabella, O., Smets, A. H., “Highly transparent modulated surface textured front electrodes for high efficiency multijunction thin-film silicon solar cells”. Progress in Photovoltaics: Research and Applications, 23(8), 949-963, 2015.

[3] Philips’Gloeilampenfabrieken, O. "A method of measuring specific resistivity and Hall effect of discs of arbitrary shape." Philips Res. Rep 13.1: 1-9, 1958

[4] Han, C. "High-Mobility TCO-Based Contacting Schemes for c-Si Solar Cells.", Diss. Delft University of Technology, 2022.