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Transparent Conductive Oxide Bi-layer as Front Contact for Multijunction Thin-film Silicon Solar Cells

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***Abstract* — Transparent conductive oxides (TCOs) are used as front electrode of thin film silicon (TF-Si) solar cells to increase power conversion efficiency. Metal oxides doped with different materials can be deployed as TCO. The preferred TCO is usually selected using a trade-off between transparency and conductivity. This work proposes a bi-layer front contact to address the limitation of this trade-off. IOH and i-ZnO are chosen as the best candidates for such architecture due to their good opto-electrical properties. A thin layer of IOH ensures good lateral conductivity and high transparency in the visible part of the solar spectrum. An additional i-ZnO layer provides minimized parasitic absorption losses along with low transverse resistivity. The best opto-electrical properties are achieved when deposition temperature and power density are set at 25 °C and 1.5 W/cm^2, 200 °C and 2 W/cm^2 for IOH and i-ZnO respectively.**

I. Introduction

The optical and electrical properties of metal oxides rely on the oxidation state of the metal, and on the amount/nature of impurities embedded in the films [1]. The simultaneous optimization of both transparency and conductivity in a single TCO layer may lead to a compromise solution. Therefore, this investigation is directed towards a front electrode with two film-stack to maximize the opto-electrical trade-off. According to H. Tan *et al.* [2], the implementation of a TCO bi-layer front contact diminishes parasitic absorption losses and contributes to increasing the efficiency of multijunction thin-film silicon (TF-Si) solar cells.

The TCOs contribute to parasitic absorption across the optical bandgap: i) in the ultraviolet (UV) region, and ii) in the near infrared (NIR) region of the solar spectrum due to free carrier absorption (FCA). The first issue can be tackled by selecting a material with high energy bandgap to reduce the absorption in the short wavelength range and meet the opto-electrical trade-off. The second issue is more challenging and thus addressed in this work through a comprehensive analysis.

This paper characterizes the optical and electrical properties of multiple TCO materials, and then focuses on the bi-layer architecture of two of these TCO films embedded in the front electrode.

II. Experimental Procedure

A. Fabrication

Four types of TCO materials are investigated in order to select the best candidates for a bi-layer TCO structure. They are hydrogenated indium-, indium tin-, intrinsic zinc- and aluminum doped zinc- oxides (IOH, ITO, i-ZnO, AZO). Corning glass (10 cm × 10 cm) is used as substrate for TCO film depositions. Each glass substrate is cleaned in acetone and isopropyl alcohol ultrasonic baths for 10 min respectively. IOH, ITO, i-ZnO and AZO layers are grown by using Radio Frequency (RF) magnetron sputtering technique. Time, power, and temperature are tuned to explore multiple deposition conditions for TCO fabrication. In the case of IOH, partial H_2O pressure is used as additional setting.

To improve film quality, TCO samples are subjected to a post-deposition annealing (PDA) treatment in an atmospheric environment. The saturated annealing time is 20 min and the annealing temperature (T_a) is varied between 130 and 250 °C.

B. Characterization

Given the complex interplay between optical and electrical properties, two main metrics are analyzed.

Electrically, the TCO is characterized by the conductivity (σ), which is directly proportional to carrier mobility (μ), free carrier concentration (N) and elementary charge (e). The Hall measurement allows to determine resistivity (ρ), measured by the so-called van der Pauw method, and then derive N and μ combined with the Hall effect [3]. Rather than conductivity, resistivity is also utilized as a device-relevant parameter to evaluate TCO electrical properties.

Optically, the sample is characterized by the absorption coefficient spectra (α). It is determined by a spectroscopic ellipsometry (SE) M-2000DI system (J.A. Woollam Co., Inc.) for single TCO layers. SE provides additional parameters such as bulk thickness (t_b), refractive index (n), extinction coefficient (k) and optical bandgap (E_g) from Tauc plot according to SE-fitted α curve. In SE analysis, the dielectric function of TCOs sample is considered homogenous in depth and modelled by combining two oscillators: Cody-Lorentz and Drude [4]. For the bi-layers, a spectrophotometric PerkinElmer Lambda 1050 system is utilized to measure transmittance (T) and reflectance (R), and α is derived by applying the Lambert-Beer law. This second method is employed due to the complexity of the SE fitting using dedicated oscillator theories on bi-layers.

III. Results and Discussion

The front contact architecture is designed by two layers (TCO_1 and TCO_2) in order to optimize the trade-off between the opto-electrical properties.

The conductivity can be improved by increasing N and/or μ. However, there is an upper limit for σ due to the limitation of dopant scattering and doping efficiency. Mobility and carrier concentration are related to each other by a rule of $\mu \propto N^{-2/3}$. Therefore, a trade-off between N and μ imposes that σ cannot continuously increase [5].

Additionally, intra-band transitions within conduction band lead to parasitic free carrier absorption in the NIR region. In metals and semiconductors, FCA is modelled by the Drude oscillator theory which defines a direct dependency between α_{NIR} and N. The optimized trade-off results in TCO films with high mobility and limited free carrier concentration from both optical and electrical perspectives.

The PDA process is utilized to further enhance the opto-electrical properties of TCOs and determine a temperature optimum, where N and α_{NIR} are minimized and, μ is simultaneously maximized.

The overall thickness should not exceed 700 nm. The TCO layer deposited as the first layer of the stack on glass (TCO_1) should be sufficiently thin (100-200 nm) to guarantee good lateral conductivity for the collection of photo-generated carriers at the front contact. The layer deposited as second (TCO_2) should ensure low absorptance in the NIR region, along with good transverse electrical properties. TCO_2 should be thick enough (500-600 nm) to make it conductive.

First, each TCO material is characterized individually on glass at different deposition conditions as deposited (no PDA). Fig. 1 shows α_{NIR} (cm^{-1}) versus ρ (Ω cm) for ITO, IOH, i-ZnO and AZO. The values of the absorption coefficient at wavelength $\lambda = 1100$ nm are taken as a good indicator for FCA in NIR. The bottom-left corner of this plot (yellow triangle) represents the ideal situation for a TCO, in which α_{NIR} and ρ are below 10^2 cm^{-1} and 10^{-2} Ω cm respectively. Among the TCOs investigated, AZO and ITO are highly absorptive, between $3 \cdot 10^3$ and $1 \cdot 10^4$ cm^{-1}, and ρ values are smaller than $1 \cdot 10^{-2}$ Ω cm. IOH samples reveal the lowest ρ values between $9 \cdot 10^{-3}$ and $7 \cdot 10^{-3}$ Ω cm. Even if the characteristics of i-ZnO are widely spread, it underlines the poor conductivity of such intrinsic material, with the lowest α_{NIR} values.

Fig. 2 illustrates the relation between μ and N. The top-left corner (yellow triangle) represents the ideal electrical trade-off: μ above 30 cm^2/Vs and N below 10^{19} cm^{-3}. The order of magnitude of N is between 10^{20} and 10^{21} cm^{-3} for IOH, ITO and AZO samples, whereas it is between 10^{18} and 10^{19} cm^{-3} for i-ZnO films. In terms of μ, IOH stands out with values above 30 cm^2/Vs in as-deposited condition.

Characterized mono-film TCOs samples do not meet the optimal trade-off indicated in the yellow region in Fig. 1 and Fig. 2. Hence, high transparency and high conductivity are suggested to be optimized separately in a bi-layer front contact

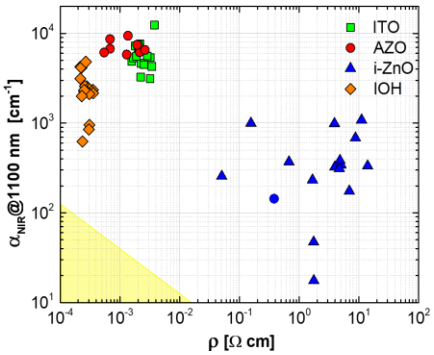

Fig. 1. Absorption coefficient @ $\lambda = 1100$ nm vs resistivity of processed TCOs. Absorption coefficient is calculated through SE.

design. Intrinsic ZnO appears to be the most suitable TCO in terms of low FCA losses, while IOH clearly achieves the lowest ρ values of all TCOs examined, by preserving high free carrier mobility. Therefore, IOH is selected as TCO_1 and i-ZnO as TCO_2 in the bi-layer structure.

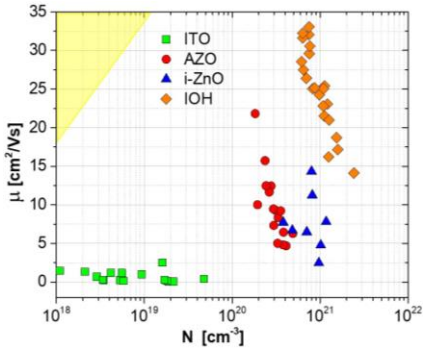

Fig. 2. Mobility vs free carrier concentration of processed TCOs.

A. TCO_1: IOH deposition conditions and PDA

Once the IOH material has been selected as the best candidate for TCO_1 in the bi-layer structure, the deposition conditions are refined using a power density window between 1.3 and 1.8 W/cm^2, and the effect of PDA on α_{NIR}, μ and N is explored. Deposition temperature, H_2O partial pressure and thickness are set at 25 °C, $3 \cdot 10^{-5}$ mbar and 200 nm respectively. Deposited samples are then annealed at 130, 180, 200 and 250 °C.

Fig. 3 shows how the progressive increase of annealing temperature rapidly decreases α_{NIR} and N values, and displays a simultaneous μ gain. A plateau occurs at 200 °C, representing the optimized PDA temperature for IOH. Higher T_a might cause hydrogen atoms effusion during the growth of the IOH film, leading to a deterioration of the film quality. Power density is chosen to be 1.5 W/cm^2 on account of the best opto-electrical trade-off seen in Fig. 3.

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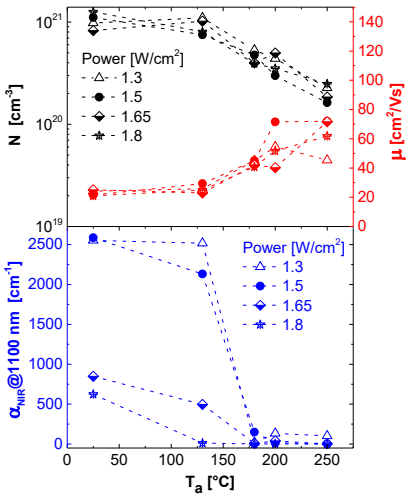

Fig. 3. Processed IOHs at different power densities (W/cm^2): mobility and carrier concentration vs annealing temperature (plot above); absorption coefficient @ λ = 1100 vs annealing temperature (plot below).

B. *TCO_2: i-ZnO deposition conditions*

The deposition conditions for i-ZnO are dependent on TCO_1. For instance, high power may damage the IOH surface due to ion bombardment. Additionally, i-ZnO cannot be deposited at higher temperature than the optimal T_a, since it would have negative effect on TCO_1's opto-electrical properties. Deposition temperature, thickness and power density are set at 200 °C, 500 nm and 2 W/cm^2 respectively.

C. *Towards bi-layer architecture*

To investigate the opto-electrical metrics of the bi-layer design, 8 samples are processed: 6 bi-layers and 2 IOH films as reference. The IOH thicknesses are 180 and 100 nm. Each IOH thickness is then combined with i-ZnO processed with varying thicknesses of 600, 500 and 427 nm.

Fig. 4 shows the shift of opto-electrical properties from IOH references (orange) to bi-layer samples (blue). Bi-layer samples further minimize the FCA losses ($\alpha_{NIR} \sim 10^2$ cm^{-1}) compared to IOH films, as shown in Figure 4 (a). However, the IOH single layer is more conductive than the bi-layer design, whose ρ values are above $1 \cdot 10^{-3}$ Ω cm. Figure 4 (b) points out that high μ (above 30 cm^2/Vs) and low N (10^{19}- 10^{20} cm^{-3}) are preserved in IOH/i-ZnO structure without any PDA treatment.

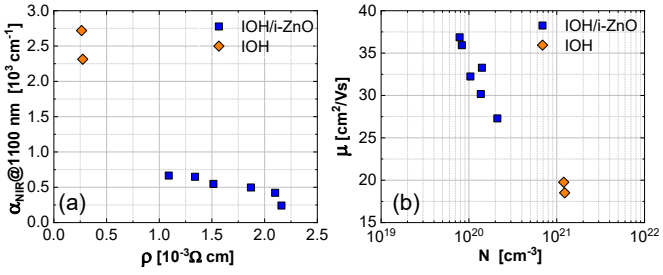

Fig. 4. (a) Absorption coefficient @ λ = 1100 nm vs resistivity of IOH/i-ZnO samples and IOH references. Absorption coefficient is calculated through R,T measurements; (b) mobility against free carrier concentration of IOH/i-ZnO samples and IOH references.

IV. Conclusions

The complex interplay between opto-electrical properties makes the optimization of single TCOs challenging. A bi-layer architecture can be an alternative solution to single TCO layer as front electrode in TF-Si application. Hence, this work designs a bi-layer structure in which IOH and i-ZnO are the best TCO candidates. This stack is able to combine good lateral conductivity and low absorption in the NIR.

The investigation is then focused on IOH and i-ZnO deposition conditions. The effect of PDA is studied on IOH. The annealing temperature (T_a) equal to 200 °C shows optimized N, μ and α_{NIR} when power density is 1.5 W/cm^2. T_a also determines a threshold for the deposition temperature of the i-ZnO material. Additionally, the power density of the i-ZnO deposition is kept under control and equal to 2 W/cm^2 due to potential damage on the IOH surface.

A bi-layer structure is fabricated using these deposition conditions, combining several thicknesses for each of the layers. This makes it possible to range the overall thickness and investigate opto-electrical metrics on multiple samples.

Results exhibit α_{NIR} between 2.5 and $7 \cdot 10^2$ cm^{-1}, and ρ between $1 \cdot 10^{-3}$ Ω cm and $2.5 \cdot 10^{-3}$ Ω cm. The majority of μ values are above 30 cm^2/Vs and N is in the range of 10^{19} - 10^{20} cm^{-3} without any post deposited annealing (PDA) treatment. Further improvements on the opto-electrical trade-off can be then achieved by including PDA in the fabrication of the bi-layer structure. Preliminary findings show μ up to 103 cm^2/Vs for annealed bilayers.

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