

Comparison Between Research-Grade and Commercially Available SnO₂ for Thin-Film CdTe Solar Cells

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X. Li, J. Pankow, B. To, and T. Gessert
National Renewable Energy Laboratory

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COMPARISON BETWEEN RESEARCH-GRADE AND COMMERCIALY AVAILABLE SnO₂ FOR THIN-FILM CdTe SOLAR CELLS

Xiaonan Li, Joel Pankow, Bobby To, and Timothy Gessert
National Renewable Energy Laboratory (NREL), 1617 Cole Boulevard, Golden, CO 80401

ABSTRACT

A comparison between research-grade, tin-oxide (SnO₂) thin films and those available from commercial sources is performed. The research-grade SnO₂ film is fabricated at NREL by low-pressure metal-organic chemical vapor deposition. The commercial SnO₂ films are Pilkington Tec 8 and Tec 15 fabricated by atmospheric-pressure chemical vapor deposition. Optical, structural, and compositional analyses are performed. From the optical analysis, an estimation of the current losses due to the SnO₂ layer and glass is provided. Our analysis indicates that the optical properties of commercial SnO₂ could be improved for PV usage.

INTRODUCTION

Fluorine-doped SnO₂ films are used extensively as transparent electrodes for thin-film photovoltaics (PV). It is a major component of the thin-film PV device. Thus, obtaining high-quality SnO₂ is critical for high-efficiency thin-film PV. This is especially true for thin-film α -Si, μ c-Si and CdTe solar cells, in which SnO₂-coated glass is used as a substrate, and the rest of the device is deposited on it. Thus, the SnO₂ properties heavily impact the junction that is grown on it.

The SnO₂-coated glass substrates used in PV devices come mainly from two sources. Research laboratories and universities generally fabricate the SnO₂ themselves. PV manufacturers (such as First Solar) use commercially available SnO₂-coated glass. However, the commercial SnO₂-coated glasses available on the market are not optimized for the PV industry. In this study, we will compare the film properties of research-grade SnO₂ films and those available from commercial sources. We will identify the differences and what could be improved. The research-grade SnO₂ film used in this study is fabricated at NREL by low-pressure metal-organic chemical vapor deposition. The commercial SnO₂ films are Pilkington Tec 8 and Tec 15, because these SnO₂-coated glass substrates are used by most PV manufacturers.

Optical, structural, and compositional analyses are performed. Our analysis indicates CdTe solar cell efficiency could be improved by optimizing the optical properties of commercial SnO₂ films.

The device results indicate that when using commercial SnO₂ substrates, the short-circuit current densities (J_{sc}) of CdTe solar cells are typically about 20 mA/cm². With research-grade SnO₂, however, a J_{sc} of ~23 mA/cm² was achieved. In this study, we present a detailed analysis to indicate where the current could be gained if the improved SnO₂ is used.

EXPERIMENTAL

The research-grade SnO₂ film is fabricated at NREL with tetramethyltin (TMT), oxygen, and bromotrifluoromethane (CBrF₃) as precursors [1]. The commercial SnO₂ films are Pilkington Tec 8 (I) and Tec 15 (II) fabricated by atmospheric-pressure chemical vapor deposition [2]. The substrate used for research-grade SnO₂ is Corning 7059, and soda-lime glass is used for commercial SnO₂.

The electrical properties of SnO₂ film were characterized with a Bio-Rad HL5500 Hall system. The total transmittance (T) and reflectance (R) spectrum were measured by a Cary 5G spectrophotometer with an integrating-sphere detector. The optical absorption (A) was calculated from $A=1-T-R$. Using the obtained optical absorption from the above characterization and AM 1.5 solar spectrum, the estimated current loss was calculated. The crystal properties and surface topography were assessed using X-ray diffraction (XRD, Scintag Model PTS) and atomic force microscopy (AFM, Autoprobe LS from Park Scientific Instruments with Si Cantilevers). SnO₂ coated glass samples were examined with a PANalytical Axios wavelength dispersive X-ray fluorescence spectrometer using a standard rhodium anode. Different conditions were utilized for various goniometer scan ranges depending on the elements of interest. Both the uncoated and tin oxide coated sides of the glass were scanned.

RESULTS AND DISCUSSIONS

Optical analysis is performed on SnO₂-coated glass substrates. Figure 1 shows the optical transmission and absorption of the research-grade and commercial (sample I, Tec 8) SnO₂ films. These two samples have similar film thickness and sheet resistance, but different optical transmission and absorption values. Of course, for commercial SnO₂-coated glass, the glass substrate contributes a large portion of the optical absorption.

Figure 2 provides a characterization that separates the effect of glass substrate and SnO₂ film. Because the coming 7059 glass has very small optical absorption, the calculation on the research-grade SnO₂ sample should be fairly accurate. The calculation on the commercial sample may indicate only a close estimate because of the high optical absorption of the soda-lime glass substrate. From Fig. 2, we can see that the glass substrate absorption of the commercial SnO₂ contributes a large portion in the long-wavelength range. In the short-wavelength range, the absorption due to the SnO₂ film is dominant.

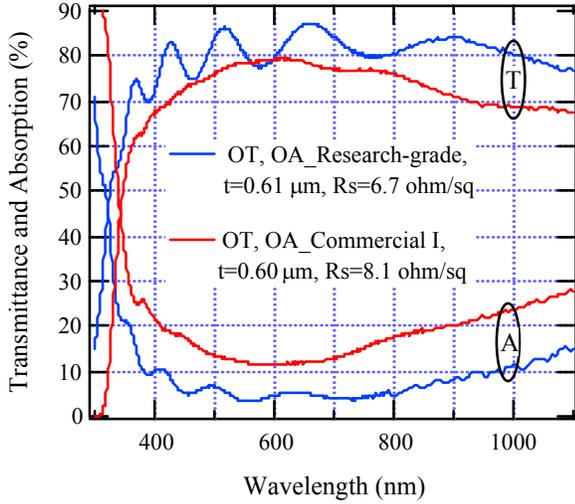


Fig. 1. Optical transmission and absorption taken from research-grade and commercial SnO₂-coated glass.

The SnO₂ film quality (e.g., structural defects and impurity levels) could cause the high optical absorption in the short wavelength, and the free-carrier scattering may account for the long-wavelength absorption. First, the optical absorption in the short-wavelength range could be due to the presence of reduced species such as Sn or SnO. In previous experiments, we found, by grazing-incidence X-ray diffraction, that annealing SnO₂ in H₂ gas will form the species Sn or SnO. Meanwhile, the optical absorption at the short-wavelength range increased significantly [3]. XRF composition analysis also found that in addition to the fluorine impurity, the commercial SnO₂ films have chlorine and other impurities, which are likely due to incorporation from CVD process. Furthermore, the saturated dopants must be considered. The classical formula for the free-carrier absorption coefficient α_f can be written as [4]:

$$\alpha_f = \frac{Nq^2\lambda^2}{m^*8\pi^2nc^3\tau} \quad (1)$$

Where N is the carrier concentration, q is electron charge, λ is photon wavelength, m^* is effective mass, n is the optical index of refraction, and τ is the relaxation time. From Eq. (1), it can be seen that the optical absorption caused by free-carrier scattering would

increase with increasing carrier concentration and photon wavelength. Thus the high carrier concentration would cause the high absorption. Figure 3 shows that for similar film thickness, the doped SnO₂ film has higher optical absorption than the undoped film in long-wavelength range. However, in the short-wavelength range, the optical absorption of the doped SnO₂ film still high, which is not due to the free-carrier effect. Therefore, we suspect that the possible existence of reduced species and a high level of impurities could be the reasons for the high absorption of the commercial SnO₂ film in the short wavelength.

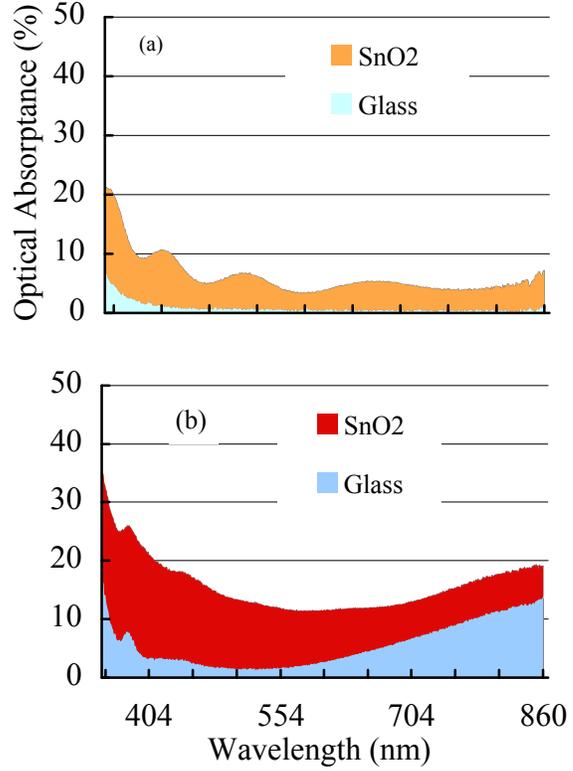


Fig. 2. Optical absorptions from (a) research-grade and (b) commercial SnO₂-coated glass (same samples as in Fig. 1). The color indicates the absorption due to different parts of the structure.

The commercial SnO₂ sample II has lower optical absorption compared to research-grade SnO₂, which is due to it being a thinner film. Thus, its sheet resistance is about 15 Ω /sq, which may be too high for CdTe thin-film solar cells. A figure of merit, $(\Phi_{TC})=T^{10}/R_s$, that can reflect both electrical and optical properties of the film is used as a comprehensive index to characterize the quality of the transparent conducting oxide (TCO) substrate. Table 1 lists electrical and optical parameters of research-grade and commercial SnO₂ films. The transmittance used for calculation Φ_{TC} in Table 1 is an average between the wavelength regions from 350 to 860 nm and taken with glass substrates. From Table 1,

we can see that considering the comprehensive index, the research-grade SnO₂-coated substrate is superior to the commercial SnO₂-coated substrate. It should be noted that for commercial SnO₂ films (with similar material properties), reducing the film thickness will increase the film transmittance, but will also increase the film sheet resistance. Thus, the comprehensive index will not change very much.

Table 1. SnO₂ Electric and Optical Properties

Sample ID	R _{sq} (Ω/sq)	*Average Transmittance (T %) (350-860 nm)	Figure of Merit ($\Phi_{TC}=T^{10}/R$) ($\times 10^{-2}$)
Research-grade SnO ₂ :F	6.7	80.17	1.636
Commercial SnO ₂ :F I	8.1	74.80	0.678
Commercial SnO ₂ :F II	14.6	79.38	0.680

*The values of average transmittance listed here are taken from film/glass structure.

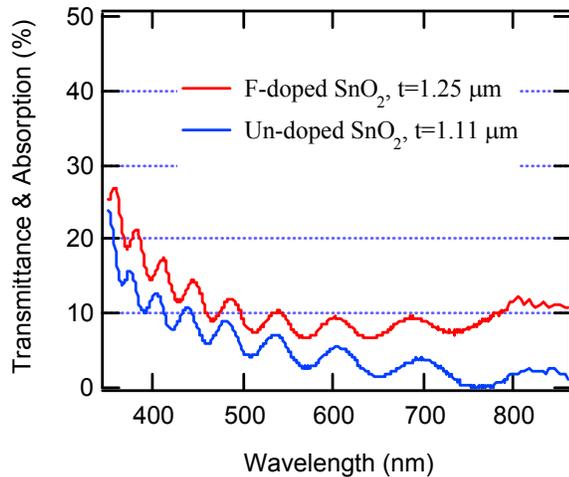


Fig. 3. Optical absorption from two research-grade SnO₂ samples.

The XRD pattern taken from the research-grade and commercial SnO₂ films are plotted in Fig. 4. Both SnO₂ films are polycrystalline. Only a single phase (tetragonal) can be identified. The commercial SnO₂ films are randomly oriented. The orientation of research-grade SnO₂ is a function of deposition temperature and fluorine doping level. At low deposition temperature and low fluorine doping condition, the SnO₂ film is randomly oriented. Otherwise, the SnO₂ film is the (200) preferred orientation [1]. The analyses on the XRD results indicate that the lattice constant of commercial SnO₂ film is larger than the bulk value, which indicates that it is under a tensile stress. The research-grade SnO₂ film shows much less stress and demonstrates high optical transparency and high electron mobility. The difference

in the film's crystal quality may partially contribute to the differences in the film's optical and electrical properties.

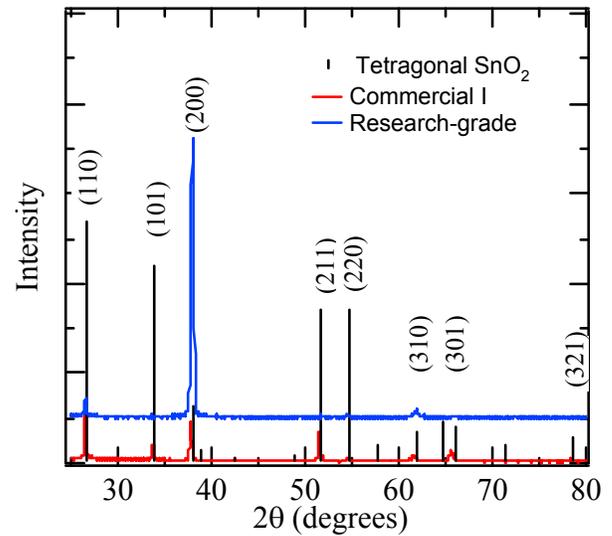


Fig. 4. XRD patterns taken from research-grade and commercial SnO₂ samples. Research-grade SnO₂ film is in (200) preferred orientation and commercial SnO₂ film is randomly oriented. The peak position of commercial SnO₂ film is moved to a lower angle, which indicates a larger lattice constant.

The possible current losses for CdTe solar cells that may be attributed to the different glass substrates and SnO₂ film coatings are listed in Table 2. Optical absorption losses were first computed from the integrated reflectance and transmittance measured on the bare glass samples and the SnO₂ films with glass substrate. We then multiplied these numbers by the AM1.5 global spectrum photon density and integrated the product in the wavelength range of 350–860 nm for CdTe and 350–1200 for μ c-Si solar cells [5]. For the commercial SnO₂ sample I, the soda-lime glass substrate causes a portion of the optical loss, but the SnO₂ film still contributes about two-thirds of the optical loss. Changing the glass substrate to a type with low optical absorption should help the device gain an additional ~ 1 mA/cm² in J_{sc}. Improving the SnO₂ material property could lead to a gain of more than 1 mA/cm² in J_{sc}. For a μ c-Si solar cell, the impact due to soda-lime glass absorption is very serious.

Furthermore, the SnO₂ quality impacts the formation of the next layer that grows on it. For the front-wall-structure CdS/CdTe device, the topography of the front TCO electrode could affect the CdS and CdTe layers that grow on it. The smooth SnO₂ surface and addition of a high-resistance buffer layer between SnO₂ and CdS will make thinner CdS possible [1]. Hence, another 1 mA/cm² would be possible depending on the final CdS layer thickness achieved.

Table 2. Possible Current Loss for the Device Due to the Optical Absorption Loss Introduced by the SnO₂-Coated Glass Substrate

Materials	Current Loss	Current Loss
	(mA/cm ²) 350-860 nm	(mA/cm ²) 350-1200 nm
Research-grade SnO ₂ /G	1.64	3.47
1.1 nm 7059 Corning glass substrate	0.19	0.31
Commercial I SnO ₂ /G	4.50	8.19
Commercial II SnO ₂ /G	2.70	6.12
3.2 mm Soda-lime glass substrate	1.70	4.17

An AFM image shows that the surface roughness (R_{rms}) of both the i-SnO₂ and SnO₂:F films depend strongly on the growth temperature and the film thickness. With a growth temperature of 500°C and a film thickness of 6000 Å, the surface roughness of SnO₂ films was about 8 nm. Table 3 lists the surface roughness of several research-grade and commercial SnO₂ samples.

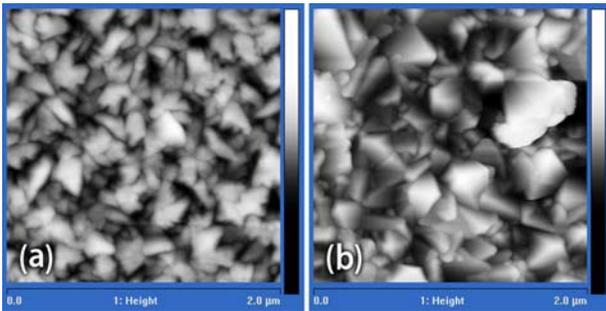


Fig. 5. AFM of (a) research-grade and (b) commercial SnO₂. Research-grade SnO₂ is fabricated at 550°C with film thickness of 1 μm, and commercial SnO₂ films have a film thickness of 0.6 μm.

Table 3. Atomic Force Microscopy Data

Sample ID	Deposition temperature (°C)	Film Thickness (μm)	Rms (nm)
Research-grade			
SnO ₂ :F-144	500	0.61	8.2
SnO ₂ :F-2374A	600	0.68	15
Commercial			
SnO ₂ /G sample I		0.6	34.8
SnO ₂ /G sample II		0.3	12.5

For research-grade SnO₂, an i-SnO₂ buffer layer is added to the structure, which further enhances its function as a front window layer for a CdTe/CdS solar cell. The device results indicate that adding an i-SnO₂ buffer layer between the SnO₂:F and CdS layers will help

to maintain the device properties. This buffer layer also helps reduce the thickness of the CdS layer. Figure 6 indicates the possible current loss as the thickness of the CdS layer increases. If smooth SnO₂ and i-SnO₂ buffer layers are used, the CdS layer thickness could be reduced considerably.

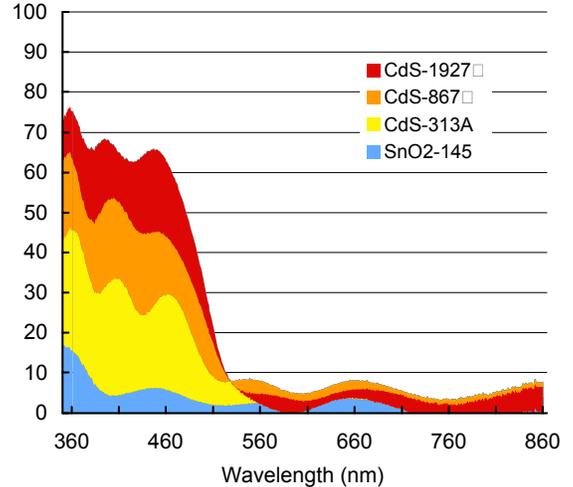


Fig. 6. Optical absorptions for three CdS samples that have different film thickness. We can see that the optical absorptions in the wavelength range of 350–550 nm are closely related to the CdS film thickness.

CONCLUSIONS

In summary, compared to commercial SnO₂ (with similar film thickness and sheet resistance), the research-grade SnO₂ has higher optical transmittance and higher electron mobility. The high optical absorption of the commercial SnO₂ substrate is due partially to the glass substrate and partially to the SnO₂ film quality. The high impurity level could contribute to the high optical absorption of commercial SnO₂. When SnO₂-coated glass used as a front conducting window layer for a CdTe solar cell, not only are high conductivity and high optical transmission required, a smooth surface also is preferred. The smooth SnO₂ surface and i-SnO₂ buffer layer will make the thin CdS layer possible. Optical absorption analyses indicate that, for commercial SnO₂, changing the glass substrate to low optical absorption glass and improving the SnO₂ film property could help a CdTe solar cell improve the photon collection and gain additional photocurrent. Furthermore, the bi-layer structure of SnO₂ with a smooth surface will make thinner CdS layers possible. With this characterization, a gain in J_{sc} by as many as 3 mA/cm² is possible with the improvement in the SnO₂-coated substrate.

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