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1 SnO₂ films elaborated by radio frequency (RF)-
2 magnetron sputtering as a potential TCOs alternative
3 for organic solar cells

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12
13 KEYWORDS: Organic solar cells; Transparent conducting oxide, Tin oxide; Reactive magnetron
14 sputtering; Thin films; Bulk heterojunction.

15

16

17 ABSTRACT

18 Transparent conducting oxides (TCOs) are crucial component of solar cells. Tin doped indium
19 oxide (ITO) is the most employed TCO, but the scarcity and high price of indium induce a search
20 for lower cost TCOs with equivalent properties as substitute. Tin dioxide (SnO_2) films have many
21 advantages, such as rich sources of material, low prices, and non-toxicity. SnO_2 films present a
22 high visible light transmittance, near-infrared light reflectivity, and excellent electrical properties.
23 They also have a higher chemical and mechanical stability compared to ITO. The aim of this work
24 is to elaborate SnO_2 films by RF-magnetron sputtering in order to use them as electrodes for
25 Organic Solar Cells (OSCs). The SnO_2 films were deposited on glass, SiO_2 and quartz substrates
26 in a mixed environment of Ar and O_2 . XRD measurements show that the as-deposited SnO_2 films
27 are polycrystalline with cassiterite tetragonal structure. SEM analysis showed that the films are
28 homogeneous, continuous, and nanostructured. The electrical resistivity and average optical
29 transmittance of the samples are about $10^{-3} \Omega\cdot\text{cm}$ and over 80%, respectively. The estimated
30 optical band gap (E_g) is around 4.0 eV while the work function of the films is around 5.0 eV. The
31 SnO_2 films are used as electrodes for inverted OSCs, using poly(3-hexylthiophene-2,5-diyl): [6,6]-
32 phenyl-C60-butyric acid methyl ester (P3HT:PC₆₀BM) as active layer. The device's open circuit
33 voltage (V_{OC}) and short circuit current density (J_{SC}) are similar to those obtained for the inverted
34 OSCs employing ITO as the same electrode. Even if the achieved power conversion efficiency is
35 lower compared to the value for the reference OSC with an ITO electrode, these results are
36 promising and place SnO_2 TCO as a potential candidate to replace ITO.

37 **1. INTRODUCTION**

38 Transparent conductive oxides (TCOs) are extensively used in modern semiconductor devices
39 such as liquid crystal displays (LCD)^{1,2}, organic light emitting diodes (OLEDs)³, touch-sensitive
40 screens⁴, photovoltaic devices^{5,6}, gas sensors^{7,8}, and smart windows⁹ due to their high electrical
41 conductivity and high optical transmission. Tin doped indium oxide (ITO) thin films are widely
42 used as TCOs because of their low resistivity (in the range of 10^{-4} Ω .cm), high light transmittance
43 in the visible region (around 90%) and multitude of wide-area deposition techniques². However,
44 indium is scarce and in high demand, making it an expensive source material¹⁰.

45 Tin dioxide (SnO₂) has received significant attention as a viable replacement for ITO due to its
46 attractive properties. SnO₂ is by nature an n-type semiconductor with a wide and direct band gap,
47 of 3.6 up to 4.0 eV at room temperature^{9,11}, and a high work function of about 5.1 eV¹¹. SnO₂ is
48 highly transparent in the visible region, chemically stable in some acidic and basic solutions¹²,
49 thermally stable in oxidizing environments at high temperature, and mechanically hard¹⁰. SnO₂
50 thin films are prepared through different deposition techniques such as DC/RF magnetron
51 sputtering^{10,13-19}, pulsed laser deposition^{20,21}, thermal evaporation^{22,23}, sol-gel²⁴⁻²⁶, spray
52 pyrolysis^{27,28}, chemical vapor deposition^{8,29,30}, and hydrothermal process^{31,32}.

53 RF-magnetron sputtering has various advantages such as good adhesion to substrates,
54 homogeneity of deposited thin films, good reproducibility, and the possibility to extend the
55 deposition technique to industrial scale³³. Therefore, it was selected as deposition technique for
56 this study. The structural, optical, and electrical properties of SnO₂ thin films deposited by RF-
57 magnetron sputtering have been investigated for a substrate temperature ranging from 100 up to
58 400°C and for an Ar/O₂ gas ratio of 8/3. The film with the lowest electrical resistivity was then
59 employed as electrode for an inverted Organic Solar Cell (OSC), and the photovoltaic properties
60 of the solar cell were studied.

61 2. Experimental section

62 2.1. Materials

63 The following chemicals were purchased: polyethylenimine (80% ethoxylated, 37 wt. % in
64 H₂O), acetone (CH₃COCH₃, ACS reagent, ≥99.5%), anhydrous 1,2-Dichlorobenzene (C₆H₄Cl₂,
65 99%), and Hellmanex™ III from Sigma-Aldrich; [6,6]-phenyl-C60-butyric acid methyl ester
66 ([60]PCBM, 99.5%) from Solenne BV; poly(3-hexylthiophene-2,5-diyl) (P3HT) from Solaris
67 Chem; molybdenum oxide (MoO₃, 99.95%) from Neyco; silver pellets (Ag, 99.99%) from RD
68 Mathis Company; ethanol absolute (C₂H₅OH, 100%), hydrochloric acid (HCl, 37%) and 2-
69 propanol (IPA) ((CH₃)₂CHOH, 100%) from VWR Chemicals; zinc powder (metals basis, -
70 140+325 mesh, 99.9%) from Alfa Aesar; chromium powder (metals basis, ≤ 5 μm, 99.8%) from
71 CERAC; AZ 4533 positive photoresist and AZ 726 metal ion free (MIF) developer from Clariant.
72 All the chemicals were directly used without further purification.

73

74 2.2. SnO₂ thin film and solar cell fabrication

75 2.2.1. SnO₂ thin films preparation

76 In order to characterize the active layer, SnO₂ thin films were deposited on oriented SiO₂ (100),
77 polished soda lime glass and synthetic quartz substrates. Prior to the deposition the substrates were
78 ultrasonically cleaned with deionized water with Hellmanex™ III, deionized water, acetone, and
79 2-propanol for 15 min each at 45°C and dried under N₂ gas flow. The thin films were deposited by
80 (RF)-magnetron sputtering using high purity (99.95%) tin (Sn) target from AJA Int. High purity
81 argon (Ar) and oxygen (O₂) were introduced into the sputtering chamber through flowmeters. The
82 Ar and O₂ flows are kept 8 sccm and 3 sccm, respectively. These gas flows were chosen based on
83 the results obtained of the study of the effect of Ar/O₂ ratio on the films resistivity (Support A).

84 The substrates were placed on a substrate holder with heating block. The working pressure was
85 kept at 3.4×10^{-3} mbar with a source-substrate distance of 25 cm and sputtering power of 50 W.
86 The deposition rate was 3 nm/min. The substrate temperatures were ranging from 100 up to 400°C
87 and an increment of 100°C is made between each treatment.

88

89 **2.2.2. Device fabrication**

90 The SnO₂ thin films deposited on glass substrates (2 cm × 2 cm) were patterned by optical
91 photolithography and wet etching (Supplementary B). The thin films were treated in UV Ozone
92 for 30 min to remove any organic residue.

93 ITO coated glass substrates (2 cm × 2 cm) were purchased from Lumtec (Taiwan). They were
94 characterized as a mean of comparison and employed for the elaboration of a reference device.
95 Prior to the deposition the ITO substrates were ultrasonically cleaned with deionized water with
96 Hellmanex™ III, deionized water, acetone, and 2-propanol for 15 min each at 45°C and dried
97 under N₂ gas flow. They were also treated in UV Ozone for 30 min to remove any organic residue.

98 The electron transport layer was prepared by spin-coating polyethylenimine (PEIE) solution
99 (0.4% in mass of PEIE in IPA) onto the SnO₂ film at 5000 rpm for 60s. The layer is then annealed
100 at 100 °C for 10 min in N₂ glove box. The P3HT:PC₆₀BM solution (donor/acceptor ratio 1:0.6)
101 was prepared by dissolving the precursors in 1,2-Dichlorobenzene (ODCB). The solution was
102 heated and stirred on a hot plate at 60°C for at least 24 h. The active layer was spin-coated from
103 the solution on the substrate in N₂ glove box at 500 rpm for 20 s then 1200 rpm for 90 s. The layer
104 is then annealed at 150 °C for 15 min. A 7 nm-thick MoO₃ layer, or hole transporting layer, and a
105 120 nm-thick Ag electrode were deposited in a single run by thermal evaporation, under high
106 vacuum (5×10^{-6} to 2×10^{-7} Torr). A metal shadow mask is used in proximity of the substrate to

107 delimit the areas to coat with the organic layer and the metal electrode. The MoO₃ and Ag layers
108 were deposited at a rate of 0.5 Å/s and 3.5 Å/s, respectively, as measured by a quartz microbalance.

109

110 **2.3.Characterization**

111 X-ray powder diffraction (XRD) patterns were collected using a Rigaku Smartlab X-ray
112 diffractometer with CuK_{α1} radiation ($\lambda = 1.54056 \text{ \AA}$) operated at 45 kV and 200 mA, in the 2θ
113 range of $15 - 70^\circ$ with a step size of 0.06° and with a scanning rate (2θ) of $0.3^\circ/\text{min}$. Phase analysis
114 was conducted using the Eva XRD software.

115 Scanning electron microscopy (SEM) was performed using a field emission scanning electron
116 microscope. Surface SEM images were recorded using a Jeol 6700 F and the cross-section SEM
117 images using a Zeiss Gemini 500. The SEM scans were recorded for 5K, 50K and 80K
118 magnification at 5 kV applied voltage for Zeiss and 7kV applied voltage for Jeol.

119 UV-Vis spectroscopy was carried out using a Perkin-Elmer Lambda 950 spectrophotometer over
120 the spectral range 300 – 1000 nm. An integrating sphere was used to collect both, specular and
121 diffuse transmittance to reduce the effect of light scattering originated from refraction and
122 reflection.

123 The thickness of the films was measured using a Veeco Dektak 150 surface profilometer. To
124 produce a step in the films, Kapton tape was applied to a small area of the SiO₂ substrate before
125 the deposition. The tape was easily removed with acetone after the deposition to reveal a step in
126 the film down to the substrate.

127 Electrical characteristics were acquired in the dark using Hall measuring equipment in the Van-
128 der Pauw's geometry, with an Ecopia HMS-5000 system, at room temperature. A direct current of

129 1 mA and a magnetic field of 0.55 T were applied. The measurements were repeated 5 times to
130 ensure reproducibility and the results were averaged to compensate deviations.

131 The films work function was measured using a Single-Point Kelvin Probe system (KP020) from
132 KP Technology in ambient air and at room temperature.

133 The current density-voltage (J-V) characteristics were recorded with a Keithley 2400 source
134 meter and a Sun 3000 Solar Simulator with the light intensity of 100 mW/cm² under AM 1.5 G
135 solar light. The active area of the measured devices was 0.12 cm², and a mask was used during the
136 measurements.

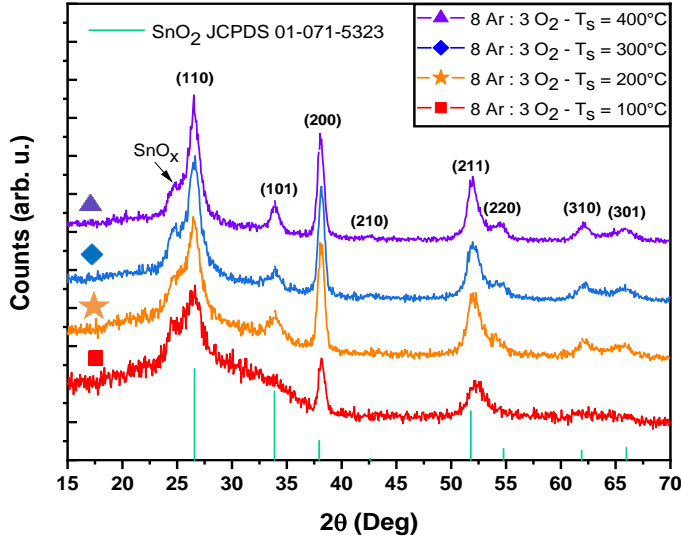
137 **3. RESULTS AND DISCUSSION**

138 **3.1. Structure and morphology**

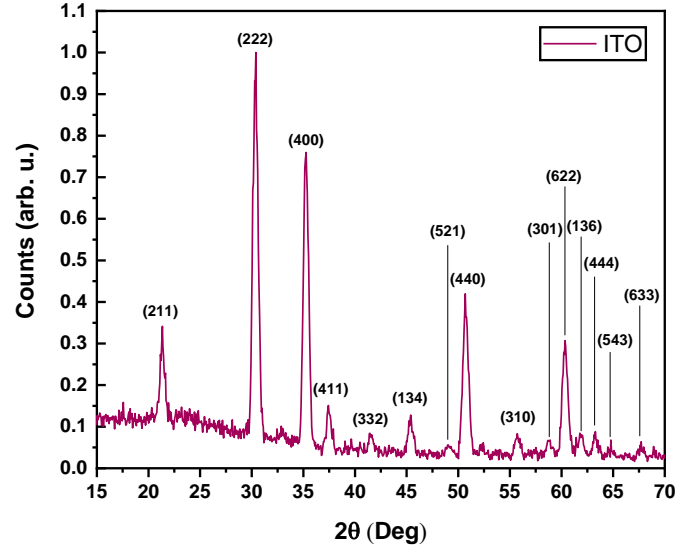
139 **3.1.1. X-Ray Diffraction**

140 The XRD patterns of the SnO₂ films deposited at different substrate temperatures T_S are shown
141 in Figure 1. All the observed diffraction peaks can be indexed according to the cassiterite tetragonal
142 structure of SnO₂ (JCPDS card n° 01-071-5323). A peak characteristic of the SnO_x phase is
143 observed at approximately 24.42° (2θ). An increase in sharpness and intensity of the XRD peaks
144 can be observed with the increase of the substrate temperature.

145



(a)



(b)

146 **Figure 1.** X-ray diffraction pattern of a) SnO₂ films deposited on glass substrates with different
 147 substrate temperatures and, b) commercial ITO. The patterns have been translated vertically for
 148 better viewing purposes.

149 The tetragonal structure lattice parameters a , b and c are defined as follows:

$$\frac{1}{d^2} = \frac{h^2 + k^2}{a^2} + \frac{l^2}{c^2}, \quad (1)$$

150 where d is the lattice spacing between two different crystallographic planes, and h , k , l the miller
 151 indexes of the reflection plane. From the results presented in Table 1 we can see that a and c are
 152 rather stable when increasing the substrate temperature T_S . Overall the lattice parameters a , b and
 153 c are slightly lower with respect to the standard values $a=b=4.737 \text{ \AA}$, $c=3.185 \text{ \AA}$ present in the
 154 literature⁹ (JCPDS card n° 01-071-5323). The lower lattice parameters and the presence of a SnO_x
 155 phase show that the SnO₂ films are probably non-stoichiometric.

156 The crystallite size D of the SnO₂ films is estimated using the Scherrer formula³⁴ (Equation 2):

$$D = \frac{K\lambda}{\beta \cos(\theta)}, \quad (2)$$

157 where D is the average crystallite size perpendicular to the reflecting planes, λ is the X-ray light
158 wavelength, β is the width of the X-ray peak on the 2θ axis, normally measured as full width at
159 half maximum (FWHM), θ is the Bragg angle, and K is the so-called Scherrer constant. K depends
160 on the crystallite shape and the size distribution, indices of the diffraction line and taken equal to
161 0.9 here. The calculated values of D (Table 1) represent estimates (discussion on the accuracy of
162 Equation (2) can be found in the literature³⁵). D slightly rises with the substrate temperature as it
163 was previously reported by Bansal et al and by Kim et al^{11,33}.

164 The amount of defects in the SnO₂ films can be determined by estimating the dislocation density
165 δ which is defined using the approach of Williamson and Smallman³⁶ (Equation 3):

$$\delta = \frac{1}{D^2}, \quad (3)$$

166 where D is the average crystallite size. As the substrate temperature increases δ decreases
167 indicating the reduction of the defects generated in the SnO₂ samples. To our knowledge no studies
168 report on the effect of substrate temperature on the dislocation density of undoped SnO₂ films
169 elaborated by (RF)-magnetron sputtering. In comparison, the commercial ITO presents a
170 dislocation density δ (Table 1) lower than all SnO₂ films which is explained by the ITO's higher
171 average crystallite size D . Therefore, the commercial ITO presumably presents fewer defects than
172 all SnO₂ films.

173 In conclusion, the XRD analysis shows that a substrate temperature of 400°C gives the best
174 crystallinity.

175

176 **Table 1.** Unit cell parameters a , b and c , unit cell volume V , crystallite size D , and dislocation
177 density δ of the SnO₂ films and commercial ITO.

Sample	$a=b$ (Å)	c (Å)	V (Å ³)	D (nm)	δ lines/ μm^2	(10 ³)
$T_S = 100^\circ\text{C}$	4.614 ± 0.011	3.063 ± 0.016	65.23 ± 0.37	11.3 4.8	\pm	7.95 ± 0.48
$T_S = 200^\circ\text{C}$	4.629 ± 0.023	3.110 ± 0.050	66.65 ± 0.86	11.7 4.7	\pm	7.29 ± 0.44
$T_S = 300^\circ\text{C}$	4.611 ± 0.010	3.140 ± 0.041	66.76 ± 0.84	12.3 4.2	\pm	6.67 ± 0.33
$T_S = 400^\circ\text{C}$	4.628 ± 0.013	3.110 ± 0.035	66.61 ± 0.67	12.6 2.7	\pm	6.28 ± 0.25
ITO	10.173 ± 0.001	10.173 ± 0.001	1052.80 ± 0.31	42.6 1.3	\pm	5.51 ± 0.33

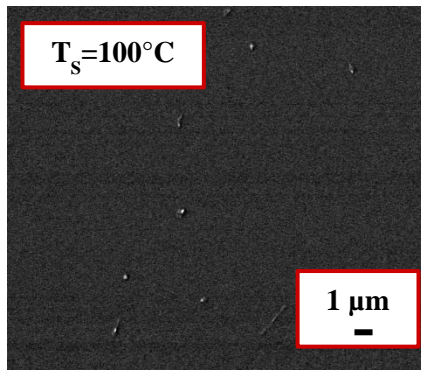
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179 **3.1.2. SEM characterization**

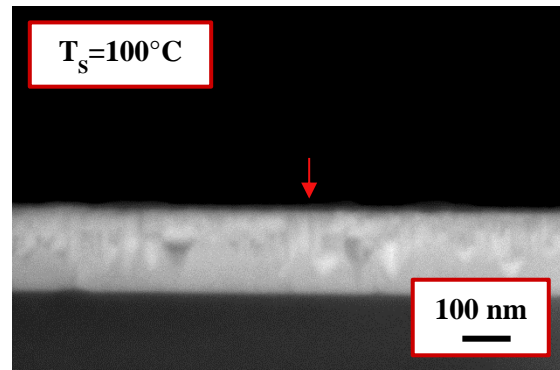
180 The SnO₂ films obtained were slightly yellowish, very clear and transparent, with smooth mirror-
181 like surface characteristic to tin dioxide³⁷. The surface and cross-section SEM images of the SnO₂
182 films deposited at different substrate temperatures are given in Figure 2. As for the surface, because
183 the grains of the films are small-sized, only some blurred SEM images can be obtained. The SEM
184 images show that as predicted by the XRD analysis the commercial ITO presents larger sized
185 grains compared to all SnO₂ films.

186 The SnO₂ films are homogenous in surface. A columnar morphology can be observed with the
187 cross-section images. The thickness of the films is homogenous and continuous throughout the
188 whole length of the films.

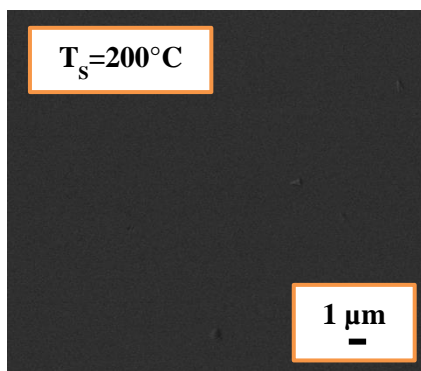
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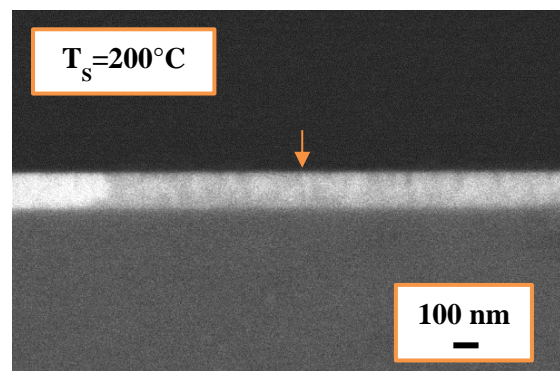
(a)



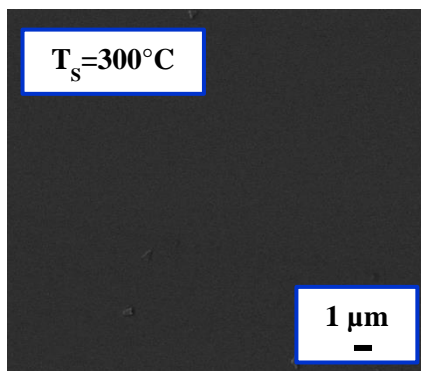
(b)



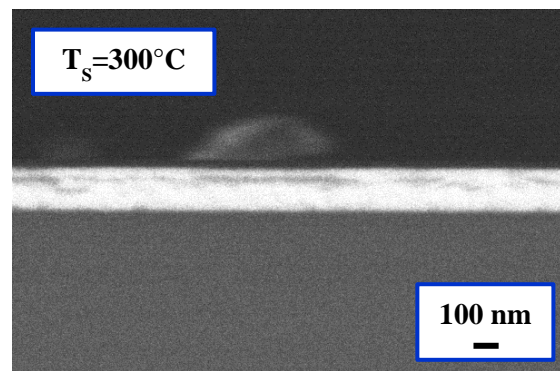
(c)



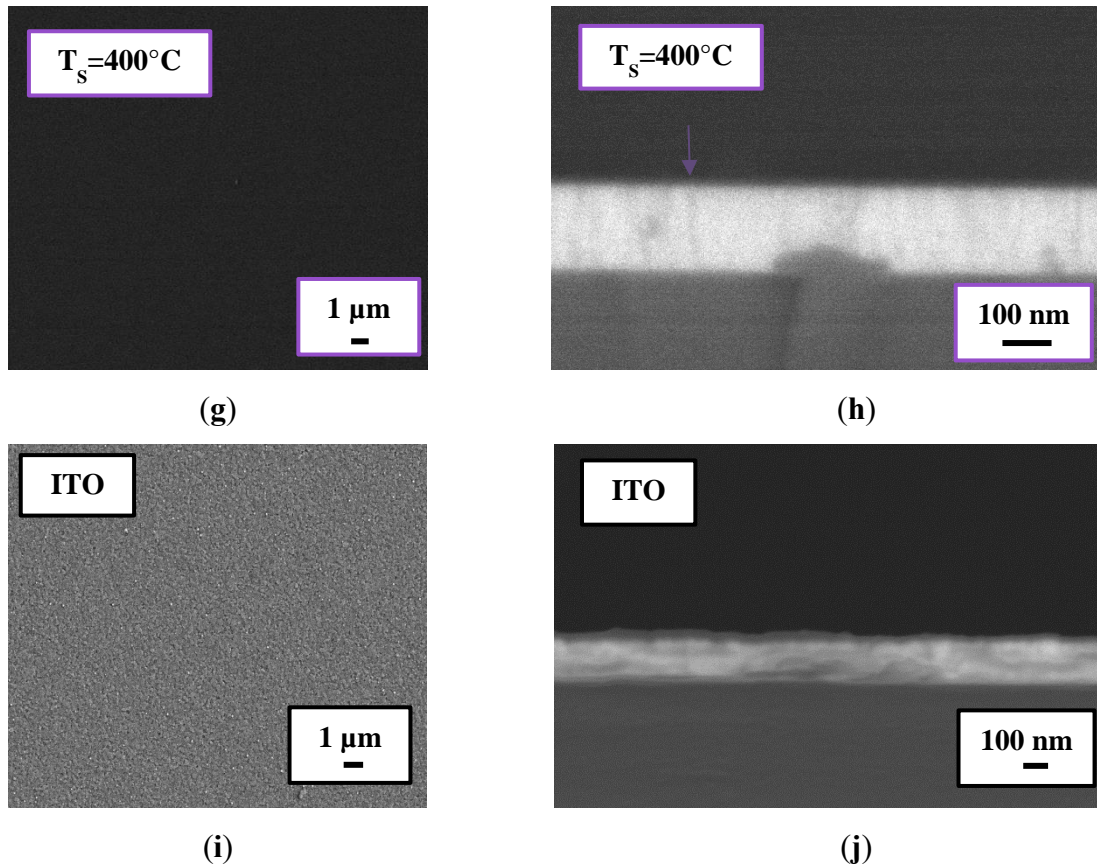
(d)



(e)



(f)



191 **Figure 2.** Surface morphology and cross-section SEM images of (a-h) SnO₂ films deposited on
 192 glass substrates for different substrate temperatures and, (i-j) commercial ITO.

193 **3.2. Optical properties**

194 Optical characteristics such as the average transmittance in the visible region and the value of
 195 the optical band gap are considered to be key features for solar cell applications. The transmittance
 196 spectra of the SnO₂ films deposited on glass substrate are displayed in Figure 3a. The average
 197 transmittance T_{avg} in the visible range increases from 83.6 up to 84.7% with the substrate
 198 temperature (Table 3). As explained by Yang et al¹⁰ a low crystallinity of the films creates inherent
 199 defects, which could cause defect scattering. On the other hand, when the crystallinity of the film
 200 is improved and the light scattering due to the inherent defect decreases an increase in the average
 201 transmittance of the film ensues, as observed for our samples. The effect of substrate temperature

202 on the average transmittance of the SnO₂ films concurs with the results in the literature^{10,12}. The
203 average transmittance of the prepared SnO₂ films are close to the values reported for undoped
204 SnO₂ films³⁷⁻³⁹ and slightly lower than the average transmittance of the commercial ITO (Table
205 2).

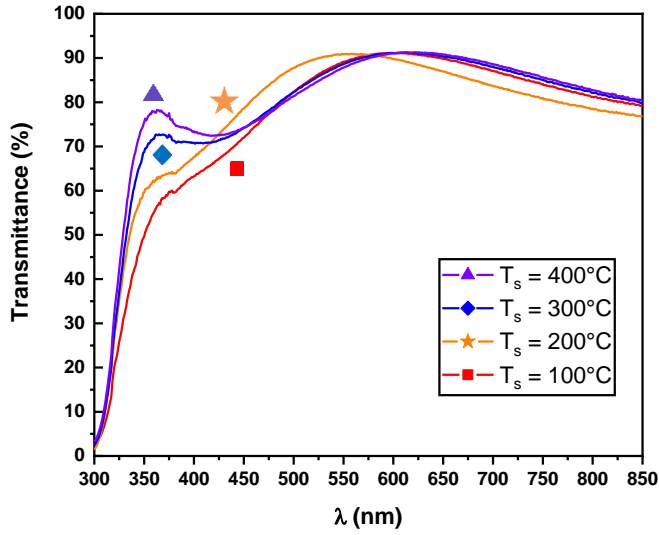
206 The incident photon energy $h\nu$ and the optical bandgap energy E_g are related through Tauc's
207 equation⁴⁰ (Equation 4):

$$(\alpha h\nu) = A(h\nu - E_g)^{\frac{1}{n}}, \quad (4)$$

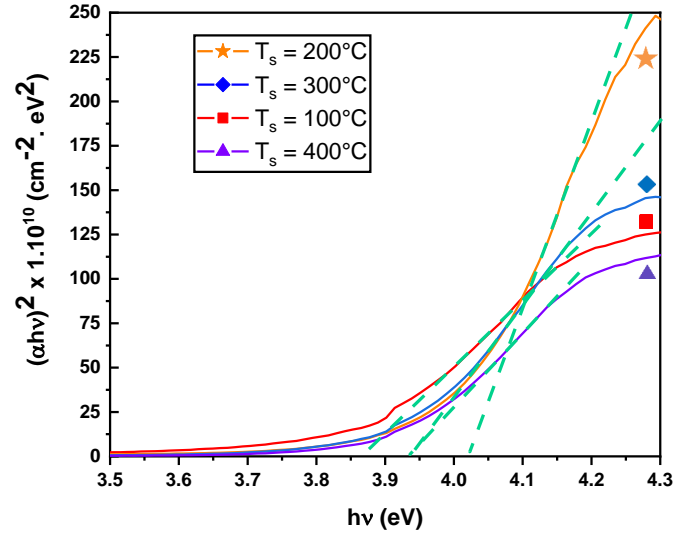
208 where α is the absorption coefficient, h is the Planck constant, $h\nu$ is the photon frequency, A a
209 constant and n is related to the type of band transition (2 or 1/2 for direct and indirect transitions,
210 respectively). The absorption coefficient α is calculated through the following relation:

$$\alpha = \frac{1}{t} \ln \left(\frac{1}{T} \right), \quad (5)$$

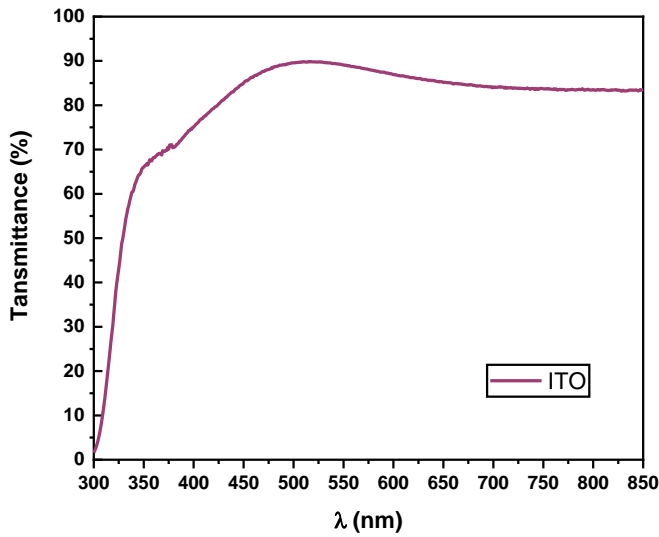
211 where T is the optical transmittance and t the thickness of the sample. The thickness t is determined
212 by profilometric analysis for each sample (Table 2). In the assumption of a direct band transition
213 for SnO₂^{10,38}, E_g was estimated by extrapolating the linear part of Tauc's plots $(\alpha h\nu)^2$ vs. $(h\nu)$ that
214 intercepts the energy axis (Figure 3b). The estimated optical band gaps are 3.86 ± 0.04 eV for T_S
215 = 100°C, 4.01 ± 0.04 eV for $T_S = 200^\circ\text{C}$, 3.93 ± 0.04 eV for $T_S = 300^\circ\text{C}$ and 3.93 ± 0.04 eV for T_S
216 = 400°C. The E_g values obtained are in agreement with the reported values for tetragonal SnO₂¹⁴.



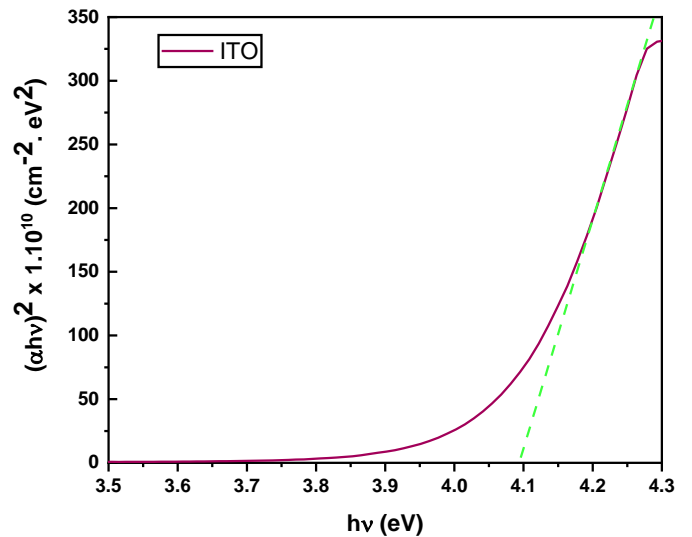
(a)



(b)



(c)



(d)

217 **Figure 3.** (a,c) UV - Visible transmittance spectrum and (b,d) Tauc's plot of the SnO₂ films for
 218 different substrate temperature and commercial ITO.

219 3.3. Electrical properties

220 The electrical resistivity ρ is connected to the carrier concentration n and Hall mobility μ by the
 221 following relation³⁹:

$$\rho = \frac{1}{ne\mu}, \quad (6)$$

222 The values of ρ , n and μ measured at room temperature for the SnO₂ films grown at various
 223 substrate temperatures on quartz are presented in Table 2. The measurements confirm the n-type
 224 nature of all prepared SnO₂ films. With the growing substrate temperature, the electrical resistivity
 225 increases from 4.45×10^{-3} up to $3.26 \Omega \cdot \text{cm}$ while the carrier concentration decreases from 6.78×10^{19}
 226 down to $1.21 \times 10^{19} \text{ cm}^{-3}$. The mobility of the free electrons also declines from 20.7 down to
 227 $1.58 \times 10^{-1} \text{ cm}^2/\text{V} \cdot \text{s}$. The effect of the substrate temperature on the resistivity of SnO₂ films observed
 228 is in agreement with the literature^{11,41,42}. The source of carriers for undoped SnO₂ is oxygen
 229 vacancies. An increment of temperature leads to a reduction in oxygen vacancies due to
 230 chemisorption of oxygen. This phenomenon results in a drop in resistivity but also in carrier
 231 concentration observed in our study¹¹. It should be noted that a resistivity of order of $10^{-3} \Omega \cdot \text{cm}$ is
 232 obtained for $T_S = 100^\circ\text{C}$, while it is usually attained for higher substrate temperature^{33,41,42}.

233 Compared to commercial ITO (Table 2), the SnO₂ film prepared at $T_S = 100^\circ\text{C}$ has a higher
 234 electrical resistivity by an order of magnitude. The difference can be explained by the higher carrier
 235 concentration n for the commercial ITO by two orders of magnitude.

236 In addition, a useful tool for comparing the performance of TCOs is the figure of merit Φ_{TC} , as
 237 defined by Haacke⁴³:

$$\Phi_{TC} = \frac{T^{10}}{R_S}, \quad (7)$$

238 where T is the optical transmittance and R_S the electrical sheet resistance of the sample. R_S is
 239 defined by the electrical resistivity ρ and the thickness d of the sample:

$$R_S = \frac{\rho}{d}, \quad (8)$$

240 The figure of merit Φ_{TC} being inversely proportional to the electrical resistivity ρ , Φ_{TC} decreases
 241 with the temperature. The estimated values of Φ_{TC} (Table 2) show that the SnO₂ film prepared at
 242 $T_S = 100^\circ\text{C}$ has the potential to be a superior TCO compared to the films elaborated at higher
 243 substrate temperatures. The resistivity of ITO being lower than the SnO₂ film prepared at $T_S =$
 244 100°C the ITO's figure of merit is higher.

245 The SnO₂ films work function (WF) must match that of the layers of the device employed in.
 246 The estimated values are presented in Table 2. For all the samples WF is around 5 eV and in
 247 agreement with the literature¹¹. The estimated values are similar to the WF of the commercial ITO.
 248 Thus, the use of a buffer layer is necessary to improve the energy level alignment with the transport
 249 levels of the donor and acceptor employed in the devices.

250

251 **Table 2.** Thickness t , average optical transmittance T_{avg} in the visible range, electrical parameters
 252 (n, ρ, μ) , figure of merit Φ_{TC} and work function WF values of the SnO₂ films and commercial ITO.

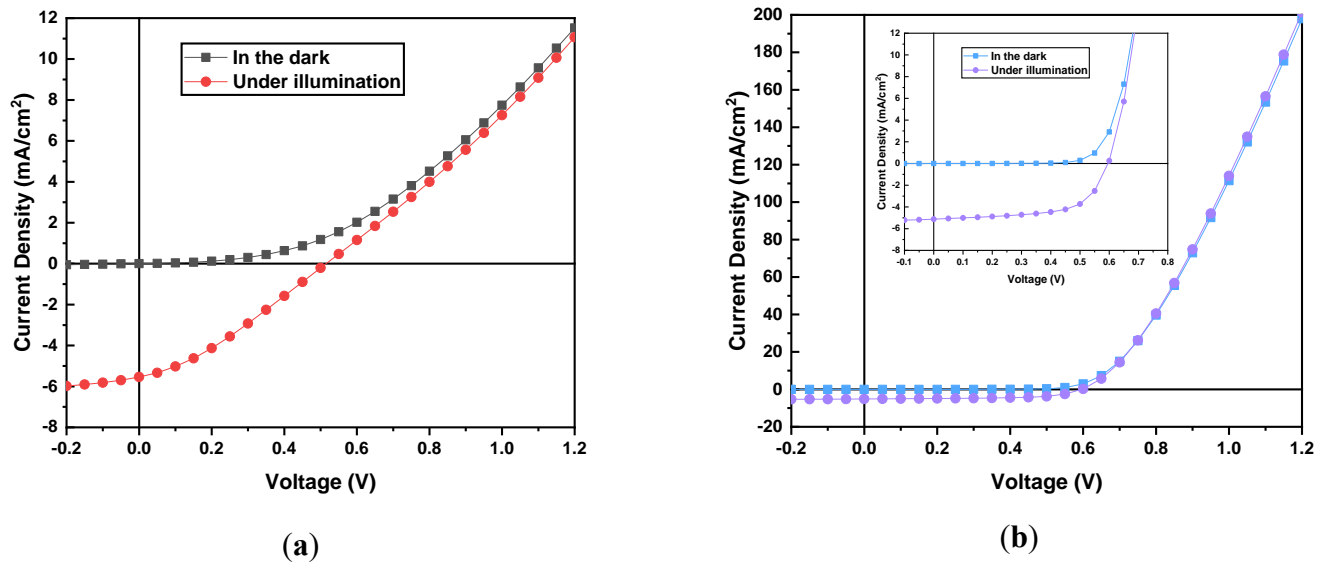
Sample	t (nm)	T_{avg} (%)	n (cm ⁻³)	ρ ($\Omega\cdot\text{cm}$)	μ (cm ² /V.s)	Φ_{TC} (Ω^{-1})	WF (eV)
$T_S = 100^\circ\text{C}$	155 ± 3	83.6	-6.78×10^{19}	4.45×10^{-3}	20.7	5.77×10^{-4}	5.02
$T_S = 200^\circ\text{C}$	160 ± 5	84.0	-5.84×10^{19}	2.75×10^{-2}	3.89	1.02×10^{-4}	5.00
$T_S = 300^\circ\text{C}$	155 ± 5	84.5	-4.22×10^{19}	2.53×10^{-1}	5.85×10^{-1}	1.14×10^{-5}	4.96
$T_S = 400^\circ\text{C}$	250 ± 10	84.7	-1.21×10^{19}	3.26	1.58×10^{-1}	1.45×10^{-6}	4.99
ITO	165 ± 5	90.5	-1.38×10^{21}	1.91×10^{-4}	26.3	1.37×10^{-3}	4.97

253

254 3.4. Photovoltaic devices

255 The requirements for an optimal TCO as replacement for ITO include a high electrical
 256 conductivity, a high transmission, and most importantly a proper work function. Therefore, the

257 SnO₂ film with the lowest electrical resistivity of $4.45 \times 10^{-3} \Omega \cdot \text{cm}$ was selected for the elaboration
 258 of an inverted OSC, using a blend of poly(3-hexylthiophene-2,5-diyl) (P3HT) and [6,6]-phenyl-
 259 C60-butyric acid methyl ester (PC₆₀BM) as active layer. A reference device with the same active
 260 layer and ITO as electrode is also elaborated. The current density–voltage (J–V) characteristics of
 261 the photovoltaic devices are shown in Figure 4 and the photovoltaic parameters are listed in Table
 262 3.



263 **Figure 4.** Photocurrent-Voltage (J-V) characteristics of (a) the SnO₂ based photovoltaic device
 264 and, (b) the ITO based photovoltaic device.

265 The SnO₂ based device has an open circuit voltage (V_{OC}) of 572 mV, a short circuit current
 266 density (J_{SC}) of 4.53 mA/cm^2 , a fill factor (FF) of 30.7%, and a power conversion efficiency (PCE)
 267 of 0.79%. The obtained J_{SC} and V_{OC} are comparable to those the ITO based device, as opposed to
 268 the FF and PCE which are lower as shown in Table 3. This difference can be explained by the
 269 impact of the series (R_S) and the shunt (R_{Sh}) resistances, and by the electrical properties of both
 270 TCOs. The SnO₂ device presents an R_{Sh} of $3.97 \times 10^3 \Omega \cdot \text{cm}^2$ compared to $4.86 \times 10^4 \Omega \cdot \text{cm}^2$ for
 271 the ITO based device indicating the presence of greater number of shunt paths in the SnO₂ based

272 device compared to the ITO one. The higher dislocation density δ of the SnO₂ film, compared to
 273 the commercial ITO ($79.5 \pm 0.48 \cdot 10^2$ lines/ μm^2 for SnO₂ compared to $5.51 \pm 0.33 \cdot 10^2$ lines/ μm^2
 274 for ITO), correlates with the assessment of made in regard of the shunt resistance. In addition, the
 275 SnO₂ based device presents a R_S of $187.3 \Omega \cdot \text{cm}^2$ about ten times higher than for the ITO based
 276 device ($17.3 \Omega \cdot \text{cm}^2$). The main effect of such a high R_S is to reduce the FF as observed. Therefore,
 277 a lower PCE for the SnO₂ based device compared to the ITO based device is observed and could
 278 be attributed to the lower electrical properties for SnO₂ compared to ITO and therefore to the lower
 279 factor of merit Φ_{TC} . Nonetheless the SnO₂ based device is functional with a high V_{OC} . And to our
 280 knowledge no undoped SnO₂ was employed as electrode in OSCs, since most of research studies
 281 on SnO₂ thin films as TCOs are focused on foreign element doping^{10,44}. These results are
 282 promising, since there is a hope to continue improving the electrical properties of SnO₂ films in
 283 order to reach efficiency close to the values obtained with ITO.

284

285 **Table 3.** Photovoltaic characteristics of the SnO₂ and ITO based photovoltaic devices.

Substrate	V_{OC} (mV)	J_{SC} (mA/cm ²)	P_m (mW)	Fill Factor (%)	PCE (%)	R_S ($\Omega \cdot \text{cm}^2$)	R_{Sh} ($\Omega \cdot \text{cm}^2$)
SnO ₂	572	5.98	7.959×10^{-5}	30.7	0.79	187.3	3.97×10^3
ITO	597	5.1	2.293×10^{-4}	61.5	1.91	17.3	4.86×10^4

286

287 4. CONCLUSION

288 Nanostructured SnO₂ films were deposited by (RF)-magnetron sputtering at different substrate
 289 temperatures for an Ar/O₂ gas flow ratio of 8/3. The structural, optical, and electrical properties of
 290 the films were investigated. The XRD profiles show that all deposited films are polycrystalline

291 and in the cassiterite tetragonal structure. The crystallinity of the films increases with the substrate
292 temperature. The average transmittance of all the films is above 80% in the visible range and the
293 optical band gap is around 4.0 eV. The SnO₂ film prepared at 100°C presents the lowest resistivity
294 of $4.45 \times 10^{-3} \Omega \cdot \text{cm}$ with a carrier concentration of $6.78 \times 10^{19} \text{ cm}^{-3}$. The transmittance, optical
295 bandgap, and work function SnO₂ films are similar to those of the commercial ITO. This leads to
296 a figure of merit (Haacke) in the same range for both SnO₂ and ITO. The SnO₂ based device
297 presents a J_{SC} and a V_{OC} values close to those obtained with the ITO based device, which is very
298 promising. Indeed, the PCE of the OSC is lower for SnO₂ than ITO only because the fill factor is
299 lower. Therefore, it is still possible to continue improving the electrical properties of the SnO₂
300 films by improving the surface quality (to raise the shunt resistance) and the conductivity (to lower
301 the series resistance, possibly by adding dopants in SnO₂). The fact that the energy levels of ITO
302 and SnO₂ are similar (bandgap, work function) demonstrates the potential of SnO₂ to replace ITO
303 in organic solar cells.

304

305 **Supplementary information**

306

307 The supplementary information is available free of charge. It contains additional experimental
308 details on the effect of O₂ partial pressure on the electrical properties of the elaborated SnO₂ thin
309 films by (RF)-magnetron sputtering, for a fixed substrate temperature of 100°C.

310 The supplementary information also contains a description of the method of patterning the
311 elaborated SnO₂ thin films.

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314

315 **Author Contributions**

316

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