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1 SnO<sub>2</sub> 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 ( $\text{SnO}_2$ ) films have many  
21 advantages, such as rich sources of material, low prices, and non-toxicity.  $\text{SnO}_2$  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  $\text{SnO}_2$  films by RF-magnetron sputtering in order to use them as electrodes for  
25 Organic Solar Cells (OSCs). The  $\text{SnO}_2$  films were deposited on glass,  $\text{SiO}_2$  and quartz substrates  
26 in a mixed environment of Ar and  $\text{O}_2$ . XRD measurements show that the as-deposited  $\text{SnO}_2$  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  $\text{SnO}_2$  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:PC<sub>60</sub>BM) 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  $\text{SnO}_2$  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)<sup>1,2</sup>, organic light emitting diodes (OLEDs)<sup>3</sup>, touch-sensitive  
40 screens<sup>4</sup>, photovoltaic devices<sup>5,6</sup>, gas sensors<sup>7,8</sup>, and smart windows<sup>9</sup> 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  $10^{-4}$   $\Omega$ .cm), high light transmittance  
43 in the visible region (around 90%) and multitude of wide-area deposition techniques<sup>2</sup>. However,  
44 indium is scarce and in high demand, making it an expensive source material<sup>10</sup>.

45 Tin dioxide ( $\text{SnO}_2$ ) has received significant attention as a viable replacement for ITO due to its  
46 attractive properties.  $\text{SnO}_2$  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<sup>9,11</sup>, and a high work function of about 5.1 eV<sup>11</sup>.  $\text{SnO}_2$  is  
48 highly transparent in the visible region, chemically stable in some acidic and basic solutions<sup>12</sup>,  
49 thermally stable in oxidizing environments at high temperature, and mechanically hard<sup>10</sup>.  $\text{SnO}_2$   
50 thin films are prepared through different deposition techniques such as DC/RF magnetron  
51 sputtering<sup>10,13-19</sup>, pulsed laser deposition<sup>20,21</sup>, thermal evaporation<sup>22,23</sup>, sol-gel<sup>24-26</sup>, spray  
52 pyrolysis<sup>27,28</sup>, chemical vapor deposition<sup>8,29,30</sup>, and hydrothermal process<sup>31,32</sup>.

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 scale<sup>33</sup>. Therefore, it was selected as deposition technique for  
56 this study. The structural, optical, and electrical properties of  $\text{SnO}_2$  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/ $\text{O}_2$  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 H<sub>2</sub>O), acetone (CH<sub>3</sub>COCH<sub>3</sub>, ACS reagent, ≥99.5%), anhydrous 1,2-Dichlorobenzene (C<sub>6</sub>H<sub>4</sub>Cl<sub>2</sub>,  
65 99%), and Hellmanex™ III from Sigma-Aldrich; [6,6]-phenyl-C<sub>60</sub>-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 (MoO<sub>3</sub>, 99.95%) from Neyco; silver pellets (Ag, 99.99%) from RD  
68 Mathis Company; ethanol absolute (C<sub>2</sub>H<sub>5</sub>OH, 100%), hydrochloric acid (HCl, 37%) and 2-  
69 propanol (IPA) ((CH<sub>3</sub>)<sub>2</sub>CHOH, 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. SnO<sub>2</sub> thin film and solar cell fabrication

#### 75 2.2.1. SnO<sub>2</sub> thin films preparation

76 In order to characterize the active layer, SnO<sub>2</sub> thin films were deposited on oriented SiO<sub>2</sub> (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 N<sub>2</sub> 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 (O<sub>2</sub>) were introduced into the sputtering chamber through flowmeters. The  
82 Ar and O<sub>2</sub> 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/O<sub>2</sub> 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  $3.4 \times 10^{-3}$  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 SnO<sub>2</sub> 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 N<sub>2</sub> 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 SnO<sub>2</sub> film at 5000 rpm for 60s. The layer is then annealed  
100 at 100 °C for 10 min in N<sub>2</sub> glove box. The P3HT:PC<sub>60</sub>BM 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 N<sub>2</sub> 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 MoO<sub>3</sub> 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 ( $5 \times 10^{-6}$  to  $2 \times 10^{-7}$  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 MoO<sub>3</sub> 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<sub>α1</sub> radiation ( $\lambda = 1.54056 \text{ \AA}$ ) operated at 45 kV and 200 mA, in the  $2\theta$   
113 range of  $15 - 70^\circ$  with a step size of  $0.06^\circ$  and with a scanning rate ( $2\theta$ ) 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 SiO<sub>2</sub> 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  $100 \text{ mW/cm}^2$  under AM 1.5 G  
135 solar light. The active area of the measured devices was  $0.12 \text{ cm}^2$ , and a mask was used during the  
136 measurements.

### 137 **3. RESULTS AND DISCUSSION**

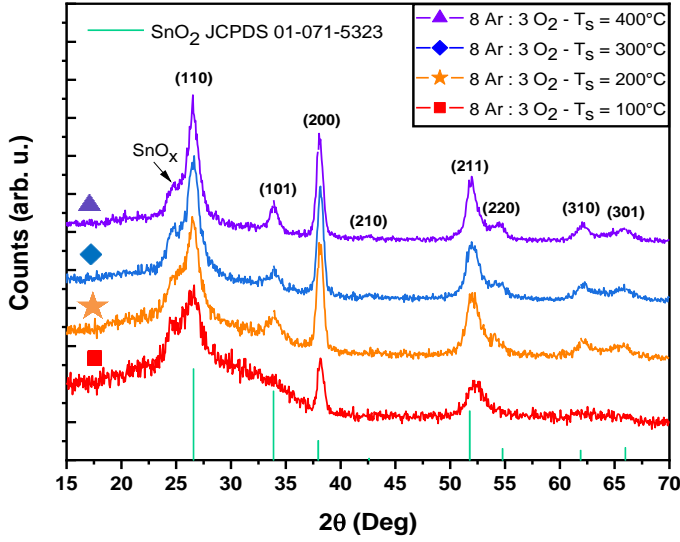
#### 138 **3.1. Structure and morphology**

##### 139 **3.1.1. X-Ray Diffraction**

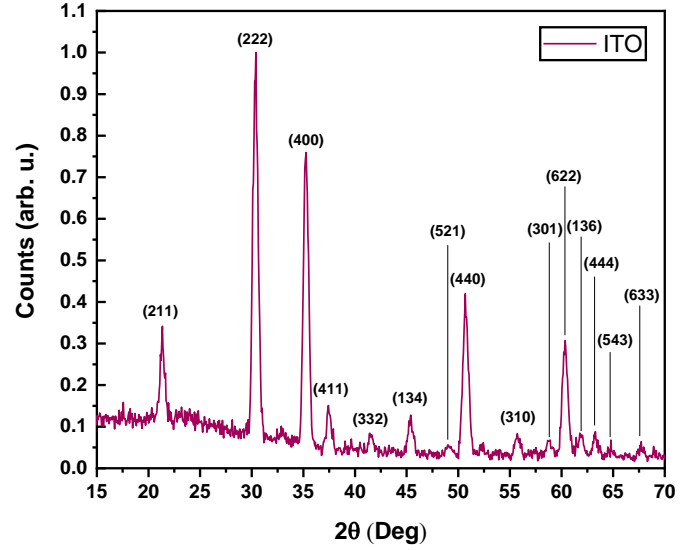
140 The XRD patterns of the  $\text{SnO}_2$  films deposited at different substrate temperatures  $T_S$  are shown  
141 in Figure 1. All the observed diffraction peaks can be indexed according to the cassiterite tetragonal  
142 structure of  $\text{SnO}_2$  (JCPDS card n° 01-071-5323). A peak characteristic of the  $\text{SnO}_x$  phase is  
143 observed at approximately  $24.42^\circ$  ( $2\theta$ ). 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) SnO<sub>2</sub> 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  $a=b=4.737$  Å,  $c=3.185$  Å present in the  
 154 literature<sup>9</sup> (JCPDS card n° 01-071-5323). The lower lattice parameters and the presence of a SnO<sub>x</sub>  
 155 phase show that the SnO<sub>2</sub> films are probably non-stoichiometric.

156 The crystallite size  $D$  of the SnO<sub>2</sub> films is estimated using the Scherrer formula<sup>34</sup> (Equation 2):

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

157 where  $D$  is the average crystallite size perpendicular to the reflecting planes,  $\lambda$  is the X-ray light  
158 wavelength,  $\beta$  is the width of the X-ray peak on the  $2\theta$  axis, normally measured as full width at  
159 half maximum (FWHM),  $\theta$  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 literature<sup>35</sup>).  $D$  slightly rises with the substrate temperature as it  
163 was previously reported by Bansal et al and by Kim et al<sup>11,33</sup>.

164 The amount of defects in the SnO<sub>2</sub> films can be determined by estimating the dislocation density  
165  $\delta$  which is defined using the approach of Williamson and Smallman<sup>36</sup> (Equation 3):

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

166 where  $D$  is the average crystallite size. As the substrate temperature increases  $\delta$  decreases  
167 indicating the reduction of the defects generated in the SnO<sub>2</sub> samples. To our knowledge no studies  
168 report on the effect of substrate temperature on the dislocation density of undoped SnO<sub>2</sub> films  
169 elaborated by (RF)-magnetron sputtering. In comparison, the commercial ITO presents a  
170 dislocation density  $\delta$  (Table 1) lower than all SnO<sub>2</sub> 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 SnO<sub>2</sub> 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  $\delta$  of the SnO<sub>2</sub> films and commercial ITO.

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

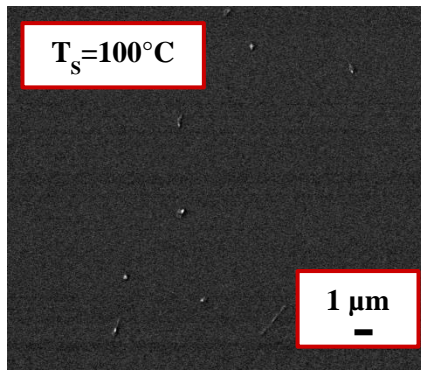
178

### 179 **3.1.2. SEM characterization**

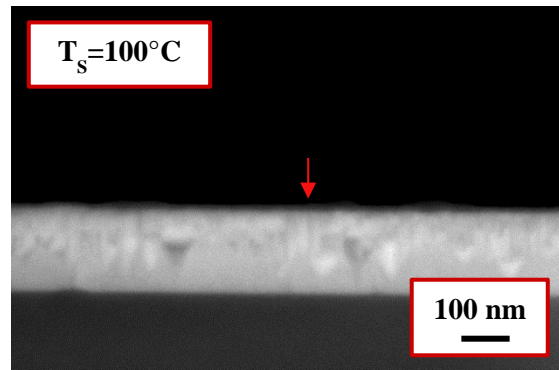
180 The SnO<sub>2</sub> films obtained were slightly yellowish, very clear and transparent, with smooth mirror-  
181 like surface characteristic to tin dioxide<sup>37</sup>. The surface and cross-section SEM images of the SnO<sub>2</sub>  
182 films deposited at different substrate temperatures are 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 SnO<sub>2</sub> films.

186 The SnO<sub>2</sub> 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.

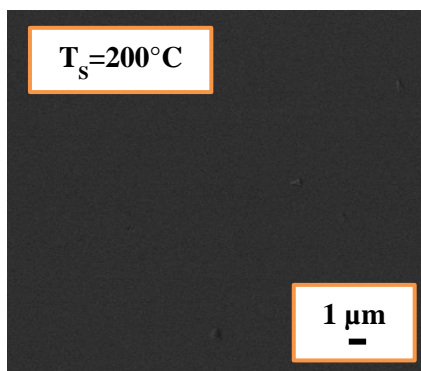
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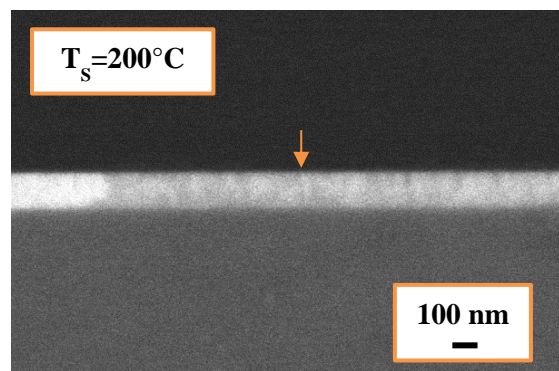
(a)



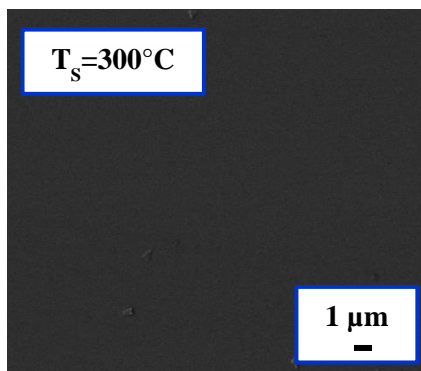
(b)



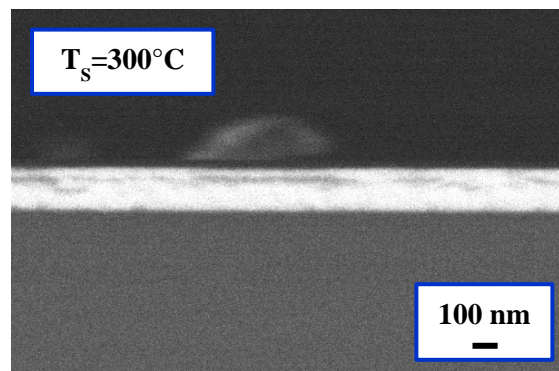
(c)



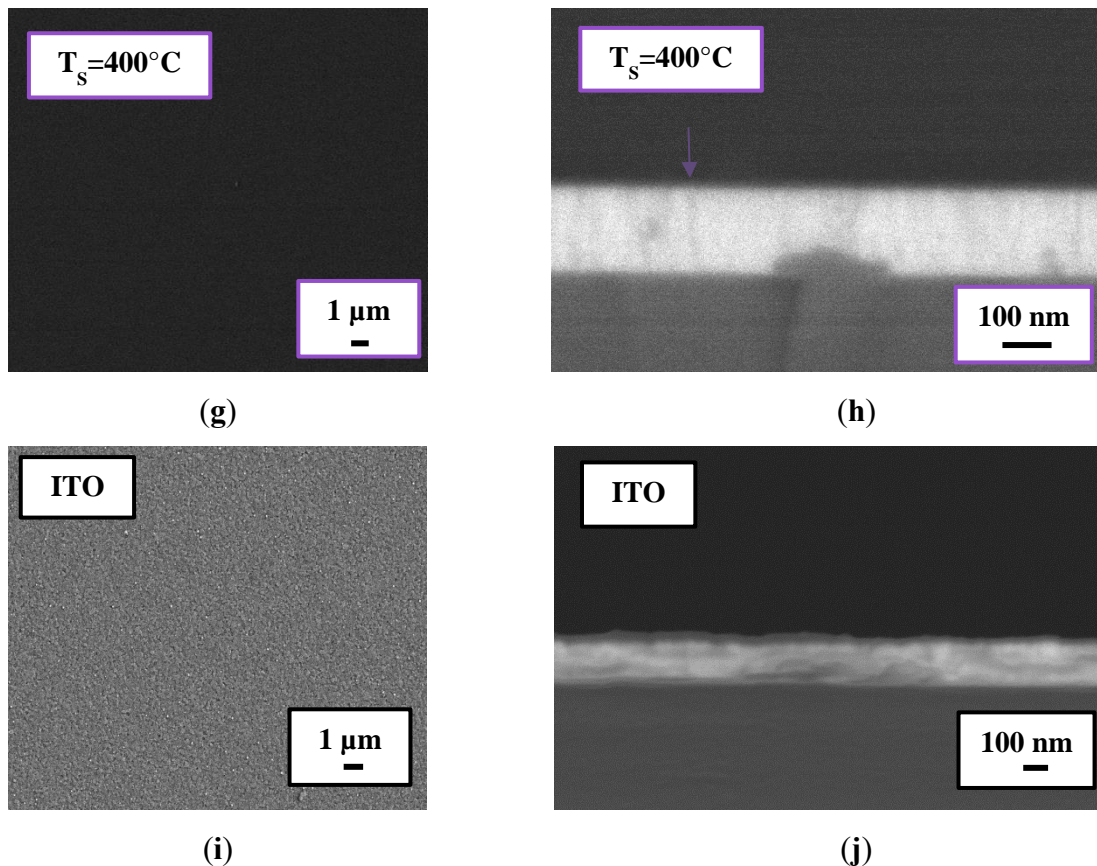
(d)



(e)



(f)



191 **Figure 2.** Surface morphology and cross-section SEM images of (a-h) SnO<sub>2</sub> 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 SnO<sub>2</sub> 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 al<sup>10</sup> 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 SnO<sub>2</sub> films concurs with the results in the literature<sup>10,12</sup>. The  
203 average transmittance of the prepared SnO<sub>2</sub> films are close to the values reported for undoped  
204 SnO<sub>2</sub> films<sup>37-39</sup> and slightly lower than the average transmittance of the commercial ITO (Table  
205 2).

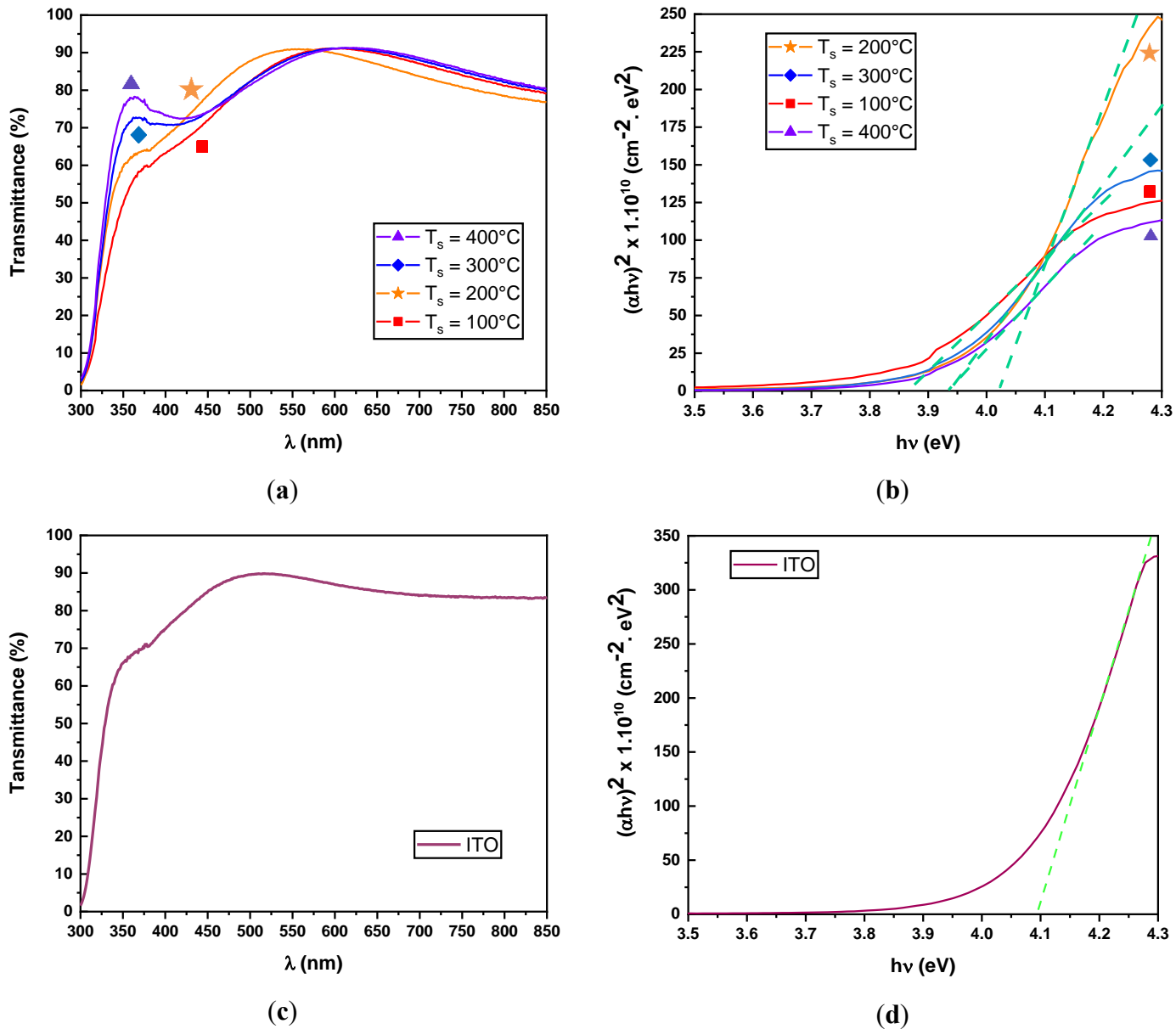
206 The incident photon energy  $h\nu$  and the optical bandgap energy  $E_g$  are related through Tauc's  
207 equation<sup>40</sup> (Equation 4):

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

208 where  $\alpha$  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  $\alpha$  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 SnO<sub>2</sub><sup>10,38</sup>,  $E_g$  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  $3.86 \pm 0.04$  eV for  $T_S$   
215 = 100°C,  $4.01 \pm 0.04$  eV for  $T_S = 200$ °C,  $3.93 \pm 0.04$  eV for  $T_S = 300$ °C and  $3.93 \pm 0.04$  eV for  $T_S$   
216 = 400°C. The  $E_g$  values obtained are in agreement with the reported values for tetragonal SnO<sub>2</sub><sup>14</sup>.



217 **Figure 3.** (a,c) UV - Visible transmittance spectrum and (b,d) Tauc's plot of the SnO<sub>2</sub> films for  
 218 different substrate temperature and commercial ITO.

### 219 3.3. Electrical properties

220 The electrical resistivity  $\rho$  is connected to the carrier concentration  $n$  and Hall mobility  $\mu$  by the  
 221 following relation<sup>39</sup>:

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

222 The values of  $\rho$ ,  $n$  and  $\mu$  measured at room temperature for the SnO<sub>2</sub> 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 SnO<sub>2</sub> films. With the growing substrate temperature, the electrical resistivity  
 225 increases from  $4.45 \times 10^{-3}$  up to  $3.26 \Omega \cdot \text{cm}$  while the carrier concentration decreases from  $6.78 \times 10^{19}$   
 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 SnO<sub>2</sub> films observed  
 228 is in agreement with the literature<sup>11,41,42</sup>. The source of carriers for undoped SnO<sub>2</sub> 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 study<sup>11</sup>. 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<sup>33,41,42</sup>.

233 Compared to commercial ITO (Table 2), the SnO<sub>2</sub> 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  $\Phi_{TC}$ , as  
 237 defined by Haacke<sup>43</sup>:

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

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  $\rho$  and the thickness  $d$  of the sample:

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



240 The figure of merit  $\Phi_{TC}$  being inversely proportional to the electrical resistivity  $\rho$ ,  $\Phi_{TC}$  decreases  
 241 with the temperature. The estimated values of  $\Phi_{TC}$  (Table 2) show that the SnO<sub>2</sub> 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 SnO<sub>2</sub> film prepared at  $T_S =$   
 244  $100^\circ\text{C}$  the ITO's figure of merit is higher.

245 The SnO<sub>2</sub> 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 literature<sup>11</sup>. 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  $(n, \rho, \mu)$ , figure of merit  $\Phi_{TC}$  and work function WF values of the SnO<sub>2</sub> films and commercial ITO.

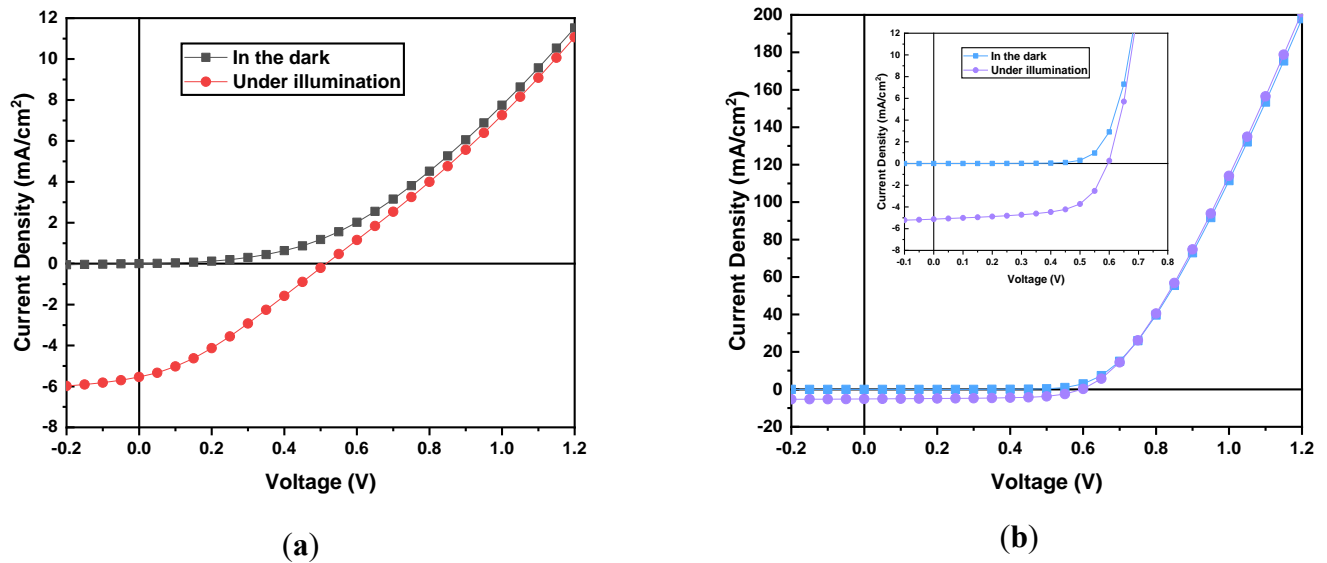
Sample	$t$ (nm)	$T_{avg}$ (%)	$n$ (cm <sup>-3</sup> )	$\rho$ ( $\Omega\cdot\text{cm}$ )	$\mu$ (cm <sup>2</sup> /V.s)	$\Phi_{TC}$ ( $\Omega^{-1}$ )	WF (eV)
$T_S = 100^\circ\text{C}$	$155 \pm 3$	83.6	$-6.78 \times 10^{19}$	$4.45 \times 10^{-3}$	20.7	$5.77 \times 10^{-4}$	5.02
$T_S = 200^\circ\text{C}$	$160 \pm 5$	84.0	$-5.84 \times 10^{19}$	$2.75 \times 10^{-2}$	3.89	$1.02 \times 10^{-4}$	5.00
$T_S = 300^\circ\text{C}$	$155 \pm 5$	84.5	$-4.22 \times 10^{19}$	$2.53 \times 10^{-1}$	$5.85 \times 10^{-1}$	$1.14 \times 10^{-5}$	4.96
$T_S = 400^\circ\text{C}$	$250 \pm 10$	84.7	$-1.21 \times 10^{19}$	3.26	$1.58 \times 10^{-1}$	$1.45 \times 10^{-6}$	4.99
ITO	$165 \pm 5$	90.5	$-1.38 \times 10^{21}$	$1.91 \times 10^{-4}$	26.3	$1.37 \times 10^{-3}$	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 SnO<sub>2</sub> 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 (PC<sub>60</sub>BM) 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 SnO<sub>2</sub> based photovoltaic device  
 264 and, (b) the ITO based photovoltaic device.

265 The SnO<sub>2</sub> based device has an open circuit voltage ( $V_{OC}$ ) of 572 mV, a short circuit current  
 266 density ( $J_{SC}$ ) of  $4.53 \text{ mA/cm}^2$ , 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 SnO<sub>2</sub> 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 SnO<sub>2</sub> based

272 device compared to the ITO one. The higher dislocation density  $\delta$  of the SnO<sub>2</sub> film, compared to  
 273 the commercial ITO ( $79.5 \pm 0.48 \cdot 10^2$  lines/ $\mu\text{m}^2$  for SnO<sub>2</sub> compared to  $5.51 \pm 0.33 \cdot 10^2$  lines/ $\mu\text{m}^2$   
 274 for ITO), correlates with the assessment of made in regard of the shunt resistance. In addition, the  
 275 SnO<sub>2</sub> 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 SnO<sub>2</sub> based device compared to the ITO based device is observed and could  
 278 be attributed to the lower electrical properties for SnO<sub>2</sub> compared to ITO and therefore to the lower  
 279 factor of merit  $\Phi_{TC}$ . Nonetheless the SnO<sub>2</sub> based device is functional with a high  $V_{OC}$ . And to our  
 280 knowledge no undoped SnO<sub>2</sub> was employed as electrode in OSCs, since most of research studies  
 281 on SnO<sub>2</sub> thin films as TCOs are focused on foreign element doping<sup>10,44</sup>. These results are  
 282 promising, since there is a hope to continue improving the electrical properties of SnO<sub>2</sub> films in  
 283 order to reach efficiency close to the values obtained with ITO.

284

285 **Table 3.** Photovoltaic characteristics of the SnO<sub>2</sub> and ITO based photovoltaic devices.

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

286

#### 287 4. CONCLUSION

288 Nanostructured SnO<sub>2</sub> films were deposited by (RF)-magnetron sputtering at different substrate  
 289 temperatures for an Ar/O<sub>2</sub> 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 SnO<sub>2</sub> 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 SnO<sub>2</sub> films are similar to those of the commercial ITO. This leads to  
296 a figure of merit (Haacke) in the same range for both SnO<sub>2</sub> and ITO. The SnO<sub>2</sub> 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 SnO<sub>2</sub> than ITO only because the fill factor is  
299 lower. Therefore, it is still possible to continue improving the electrical properties of the SnO<sub>2</sub>  
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 SnO<sub>2</sub>). The fact that the energy levels of ITO  
302 and SnO<sub>2</sub> are similar (bandgap, work function) demonstrates the potential of SnO<sub>2</sub> 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 O<sub>2</sub> partial pressure on the electrical properties of the elaborated SnO<sub>2</sub> 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 SnO<sub>2</sub> thin films.

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314

315 **Author Contributions**

316

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