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1 **Microwave plasma and rapid thermal processing of Indium-Tin oxide**
2 **thin films for enhancing their performance as transparent electrodes**

3
4 **R. Ramadan^{1,2,*}, K. Abdelhady², M. Manso-Silvan¹, V. Torres-Costa¹, and R.J. Martın-**
5 **Palma¹**

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7 ¹Departamento de Fısica Aplicada, Universidad Autonoma de Madrid, 28049 Madrid, Spain.

8 ²Physics Department, Faculty of Science, Minia University, Minia, 61519, Egypt.

9 * Corresponding author: E-mail address: rehab.ramadan@predoc.uam.es

10
11
12 **ABSTRACT**

13
14 Indium-tin oxide (ITO) is widely used as a transparent electrode for optoelectronic
15 devices given its large transparency and high conductivity. However, the particular
16 properties of this material greatly depend on the overall fabrication process. In this work,
17 we report on the effect of four different post-fabrication processes on ITO thin films
18 grown by electron beam evaporation. More specifically, the effect on the overall
19 properties of evaporated ITO thin films of microwave plasma annealing, rapid thermal
20 processing, and the two processes combined were analyzed. In particular, the
21 morphological, chemical, optical, and electrical properties of the annealed ITO thin films
22 were studied and discussed. The experimental results show that the ITO thin films can be
23 turned from opaque to transparent and their conductivity can be improved by one order
24 of magnitude depending on the particular post-fabrication process.

25
26 **Keywords:** Indium- tin oxide, microwave plasma annealing, rapid thermal processing,
27 surface morphology and optoelectronic properties.

1. INTRODUCTION

Transparent conductive oxides (TCOs) have large band gaps while their Fermi level is located in the conduction band due to free-carrier producing centers [1]. These properties make TCOs very good candidates for their use as transparent electrodes in the development of photonic and optoelectronic devices. In particular, TCOs have been used for the fabrication of organic light-emitting (OLED) devices [2], liquid crystal displays [3], electrochromic windows [4], and photovoltaic solar cells [5]. Logically, their optimum performance relies on a large optical transparency in the visible range combined with high electrical conductivity. However, although TCOs have been used for several applications during the past six decades, many unanswered questions at both the fundamental and applied levels still remain.

TCOs can be made out of p-type and n-type semiconductor materials. p-type TCOs, such as cupric oxide, have found very limited applications since the fabrication of efficient p-type TCOs remains an outstanding challenge. For instance, they have been used in bifacial and multijunction cells [6]. Accordingly, most commercially available TCOs are n-type semiconductor materials, including Sn-doped In_2O_3 (ITO) [7], Al-doped ZnO [8], and F-doped SnO_2 [9]. Among these, ITO has the best optoelectronic properties because it has high electrical conductivity and large transparency in the visible region of the spectrum, together with high reflectance in the infrared wavelength interval. Besides, this material has a wide bandgap ($E_g = 3.5 - 4.2$ eV) [10], [11].

There are many different techniques used for the deposition of ITO thin films, although most of them are based on vacuum processes such as DC magnetron sputtering [12, 13], RF magnetron sputtering [14], chemical vapor deposition [15], pulsed laser deposition [16], reactive ion plating [17], and electron beam evaporation [18]. Electron beam (e-beam) evaporation is one of the most effective techniques used for the deposition of ITO thin films with high electrical conductivity and large optical transparency. However, there are many parameters which affect the overall quality of the evaporated thin films. These include doping level, annealing process, and the factors related to the specific method of evaporation. For example, the properties of thin films grown by e-beam evaporation are affected by such parameters as the pressure in the vacuum chamber, electron beam

67 intensity, evaporation rate, etc. All these experimental variables have a notable effect on
68 the chemical composition, crystallinity, electrical conductivity, and optical properties of
69 the TCOs [18].

70

71 In this work, we present our findings regarding the study of the effect of four different
72 annealing processes on ITO thin films grown by electron beam evaporation. More
73 specifically, the effect of microwave plasma annealing (MwPA), rapid thermal annealing,
74 and these two processed combined on the overall properties of evaporated ITO thin films
75 is analyzed. In particular, the morphological, chemical, optical, and electrical properties
76 of the resulting ITO thin films are studied in detail. We aim at understanding the particular
77 mechanisms responsible for the unique combination of optical and electrical transport
78 properties of the fabricated ITO thin films.

79

80 **2. EXPERIMENTAL**

81 **2.1 Fabrication and processing techniques**

82

83 Indium tin oxide (ITO) thin films were evaporated on quartz and silicon substrates
84 ($1.5 \times 1.5 \text{ cm}^2$) by electron beam (e-beam) evaporation. The source material is indium
85 oxide/tin oxide, 90%/10%wt.%, which was purchased from the Kurt J. Lesker Company.
86 The typical base pressure was 2×10^{-7} Torr, which grew to $4\text{--}6 \times 10^{-5}$ Torr during the
87 evaporation process. The evaporation process was carried out without introducing any
88 reactive gases. The substrate was located at around 30 cm over the ITO target. The
89 emission current was 30 mA and the deposition time was 30 minutes. It is worth noticing
90 that the thin films showed a brownish tonality upon evaporation.

91

92 Rapid thermal processing (RTP) in oxygen atmosphere at 550 °C for 5 minutes was used
93 to change the overall properties of the thin films. The heating ramp until the desired
94 temperature was reached was of around 52.5°C/s and the cooling ramp was around
95 17.5°C/s. It was observed that the thin films subjected to RTP treatments showed large
96 transparency.

97

98 In addition to RTP, the effect of microwave plasma annealing (MwPA) on the ITO thin
99 films grown by evaporation was studied. The MwPA system consists of a cylindrical glass

100 vacuum chamber inserted in a stainless steel earthed hood. An open window orients the
101 Mw wave-guide to the glass cylinder, while the hood acts as a resonant cavity for the
102 solid state generated microwaves (2.45 MHz, up to 600 W). The conditions of the
103 atmosphere are controlled through an exhaust valve with a rotary pump allowing a
104 background pressure of 2×10^{-2} mbar and a common inlet for reactive and inert gasses
105 with individual gas flow meters. A grounded, refrigerated stainless steel substrate holder
106 exposes the sample horizontally to the Mw plasma. MwPA treatments were performed in
107 an argon atmosphere, with a typical pressure of 0.1 mbar for 2 minutes ON/OFF steps
108 every 60 s and at a maximum power of 600W [19].

109

110 **2.2 Characterization techniques**

111

112 The morphology of the thin films was studied by field emission scanning electron
113 microscopy (FESEM) using a Philips XL- 40FEG microscope operated at 10 kV. An
114 Energy Dispersive Spectroscopy X-ray analyzer (EDX, Inca X-sight 7558, Oxford
115 Instruments) coupled to the microscope was used to determine the elemental
116 compositions of the films.

117

118 X-ray diffraction (XRD) studies were performed using a Siemens D5000 HR
119 diffractometer in the grazing-incidence configuration using Cu-K α radiation ($\lambda=1.54$ Å),
120 a fixed incidence angle of 0.5 degrees and 2θ range from 20° to 70° with 0.04° increments
121 and 10 s accumulation time.

122

123 Optical characterization in the UV-visible range (300-850 nm) of the ITO thin films,
124 comprising transmittance (T) and reflectance (R), was carried out using a Jasco V-560
125 double-beam spectrophotometer, equipped with an integrating sphere to avoid scattering
126 losses. For this particular purpose, the ITO thin films were deposited on quartz substrates.
127 The spectral values of the optical constants (index of refraction, n , and extinction
128 coefficient, k) and the thin film thickness were determined using a program developed in
129 our laboratories based on genetic algorithms [20].

130

131 The resistance of the thin films was measured using the two point probe method. A
132 homemade cell consisting of two movable copper probes (with a diameter of 0.5 mm) and

133 a grounded copper base ($2 \times 2 \text{ cm}^2$). The distance between the two probes was 1.5 cm. The
134 measurements were carried out in a Faraday's box to shield them from external signals.
135 The electrical measurements were carried out in a Bio- Logic SP-150 potentiostat with a
136 scan rate 20 mV/s and applied potential in the 0 to 1 V range.

137

138 All the measurements were performed on samples grown onto monocrystalline Si
139 substrates except the optical measurements for which quartz substrates were used.

140

141

3. EXPERIMENTAL RESULTS

142

143 Five different sets of ITO thin films were the subject of the present study, namely (1) as-
144 deposited thin films and thin films subjected to (2) MwPA, (3) RTP, (4) MwPA plus
145 subsequent RTP (MwPA+RTP), and (5) RTP plus subsequent MwPA (RTP+MwPA).
146 The morphological, chemical, structural, optical, and electrical properties of the
147 previously described sets of thin films were studied. The results are described in the
148 following sections.

149

150 3.1 Morphology

151

152 Fig. 1 shows typical cross-sectional FESEM images of the different ITO thin films before
153 and after processing. It is evident that, in all cases, the ITO thin films show good
154 homogeneity, both from the point of view of the overall structure and thickness. Besides,
155 no cracks or other imperfections are manifested. Moreover, it is observed that the ITO
156 thin films show a granular composition. What is more, the different images show clear
157 differences in the average sizes of the nanoparticles which compose the thin films, an
158 effect which most likely has its origin in the different annealing processes. As we will see
159 below, MwPA processing has a notable effect on the crystallinity of the thin film, since
160 MwPA-processed samples present the largest particle size as determined by XRD
161 analysis. This issue will be discussed in detail in the next section.

162 From the cross-sectional FESEM images, the thickness of the various thin films was
163 determined by means of the image analysis program package Imagej [21]. The values are
164 presented in Table 1, from which it is observed that the thickness slightly decreases upon
165 annealing [10], probably due to film compaction by heat treatment. This effect might have

166 its origin in partial crystallization as will be discussed in section 3.3 (Structural
167 properties).

168

169 Table 1: Thickness of the ITO thin films before and after the different post-deposition treatments,
170 experimental values of the sheet resistance, resistivity, and 2θ ($^\circ$) for the (222) plane. FWHM and grain size
171 (D) of the ITO thin films determined from the XRD spectra.

ITO thin film name	Film Thickness (nm)	Sheet resistance (Ω/\square)	Resistivity $\times 10^{-4}$ ± 0.5 ($\Omega\cdot\text{cm}$)	2θ ($^\circ$) for the plane (222)	FWHM (radian)	Grain size (nm)
As deposited	574	84.98	48.7	32.20	3.84	2.16
MwPA	570	3.51	2.0	30.72	0.4527	18.33
RTP	566	29.24	16.5	30.70	0.4847	17.12
MwPA+RTP	540	28.02	15.1	30.75	0.5070	16.36
RTP+MwPA	518	8.30	4.3	30.73	0.5359	15.48

172

173

174 3.2 Semi-quantitative EDX microanalysis

175

176 Fig. 2 gives the elemental compositions of the different ITO thin films deposited on Si
177 (100) substrates. The experimental results confirm that, as expected, In, Sn and O are the
178 main elements of the ITO thin films. It has to be noted that the Si signal comes from the
179 substrate and the small Cr signal has its origin in the very thin layer (around 15 nm)
180 deposited by sputtering before FESEM characterization for metallization. The images
181 prove that all the films are free from contaminants and show a quite good compositional
182 homogeneity. Fig. 2(6) depicts the weight percentage of In and Sn in the different ITO
183 films, from which a slight variation of the relative content of the two elements is noted.
184 This indicates that the post-formation processes (RTP, MwPA, and their combination) do
185 not have a notable effect on the main elemental compositions. These results agree with
186 previous studies [22], [23]. Furthermore, the Sn to In ratio remains almost constant and
187 close to the doping concentration value of Sn in the source material, which is indium
188 oxide/tin oxide: 90%/10%wt.%.

189

190 3.3 Structural properties

191

192 The microstructure of the ITO thin films was studied by X-Ray Diffraction (XRD)
193 analysis. It is well known that the microstructure of the ITO thin films is extremely
194 sensitive to the specific annealing process, as previously reported [10]. Our experimental
195 results, shown in Fig. 3, allow to conclude that the as-deposited ITO thin films are
196 amorphous and only one single broad diffraction peak is observed. However, all
197 processed thin films show a well-defined crystalline structure with diffraction peaks
198 corresponding to specific crystallographic planes. In particular, the (222) plane appears
199 as the preferred orientation as demonstrated by the highest intensity of the corresponding
200 diffraction peaks. This observation is in agreement with previous works [10], [24].
201 Additionally, it is noted that annealing of ITO thin films has a notable effect on the grain
202 size (D).

203

204 In order to estimate the grain size of the ITO thin films from the preferential oriented
205 (hkl) crystal plane, the Debye Scherrer formula [25] was used:

206

$$D = \frac{0.94 \lambda}{\beta \cos \theta},$$

207 where λ is the wavelength of the X-ray radiation used ($\lambda=0.154$ nm in our case), β is the
208 measured broadening of the diffraction line at half maximum intensity (FWHM) in
209 radians, and θ is the corresponding Bragg's diffraction angle of the main peak in the XRD
210 plots. For the experimental determination of the values of D , the (222) plane was used
211 given these show the largest intensity, as pointed out before. The experimental results,
212 which are summarized in Table 1, indicate that the grain sizes for all the processed
213 samples are larger than those of the as-deposited film. Besides, thin films processed by
214 MwPA show the largest values, which was previously ascertained by FESEM analysis
215 (Section 3.1). However, it has to be noted that the XRD peaks can be widened by defects
216 and internal stress. For this reason, the mean values of D calculated by this method are
217 generally smaller than the real ones [26].

218

219 3.4 Optical properties

220

221 Fig. 4 shows typical reflectance (R) and transmittance (T) spectra of as-deposited ITO
222 thin films grown on quartz substrates. It is observed that T is negligible, while the

223 reflectance spectrum is practically flat and the values are around 20 % in the 400-850 nm
224 wavelength range. However, MwPA processing of the ITO thin films leads to notable
225 changes in the optical properties, i.e., while T remains close to zero, the values of
226 reflectance are quite low at short wavelengths but increase towards the infrared, as
227 depicted in Fig. 4. Fig. 5 portrays the R and T spectra of the ITO thin films upon RTP
228 processing, and a combination of the two previous techniques, namely MwPA+RTP and
229 RTP+MwPA. The experimental spectra confirm the high transparency of the three
230 different thin films, with an average value of T around 85% and of R about 12.
231 Additionally, it is observed that the experimental curves present a few maxima and
232 minima in the visible and near infrared wavelength regimes, due to interference effects.
233 The position of the maxima and minima remains almost constant for the three samples,
234 which confirms the small variation of the thickness of the thin films after the annealing
235 processes and also the minor variation of the chemical composition, leading to small
236 variations of the index of refraction. This particular issue will be discussed later in this
237 section. Finally, below about 400 nm a quite large absorption is clearly observed, not
238 attributable to the substrate, since quartz substrates were used for the optical
239 measurements.

240 From the experimental optical T and R spectra of the ITO thin films, the values of the
241 optical constants, refractive index (n) and extinction coefficient (k), were extracted. For
242 this purpose, a program developed in our laboratories was used [20]. Figs. 6 and 7 show
243 the spectral values of n and k , respectively, for the ITO thin films. The results show that
244 the values of n decrease with increasing wavelength for all the ITO thin films. Moreover,
245 the experimental results show that annealing leads to increased values of the index of
246 refraction for all the treated thin films. This effect is attributed to the crystalline structure
247 of the ITO thin films upon processing, as determined by XRD analysis (Section 3.3). As
248 previously indicated, the grain size increased from around 2 nm for as-deposited thin
249 films to about 16 nm for the annealed ITO thin films. As a result of the crystallinity of
250 the processed thin films, the ITO nanocrystals are more closely packed together. As such,
251 the thin films are more compact leading to a higher refractive index as a consequence of
252 the reduction of void space. These results are in agreement with previous works, such as
253 those which studied the effect of Sn doping on the properties of ITO thin films prepared
254 by e-beam evaporator [27], the effect of annealing processes on the optical constants of
255 MgF_2 thin films [28], TiO_2 nanostructured thin films [29], and annealed $\text{Cd}_{1-x}\text{Zn}_x\text{Se}$ thin

256 films (with $x = 0, 0.40$ and 1) [30]. Wherefore, the curves for the transparent films (RTP,
257 MwPA+RTP, and RTP+MwPA) show a small variation in the spectral values of n and
258 this limited variation agrees with the rather small changes in the thickness, grain size, and
259 transparency. A similar behavior for the spectral values of the extinction coefficient is
260 observed, as depicted in Fig. 7. The values of k are very small and close to zero for all the
261 samples as a consequence of the small optical absorption of the ITO films, leading to
262 large transparency in the visible range. More specifically, the three transparent thin films,
263 RTP, MwPA+RTP, and RTP+MwPA, show very low values of k , below 10^{-3} . However,
264 the opaque thin films, namely as-deposited ITO and MwPA-processed, have values of k
265 larger than 10^{-1} , as shown in Fig. 7.

266 In addition, the values of the optical band gap for the transparent ITO thin films, i.e., thin
267 films treated by RTP, MwPA+RTP and RTP+MwPA, were extracted by using Tauc plots
268 [31], as shown in Fig. 8. The absorption coefficient (α) of the films were determined from
269 optical measurements in the 400 to 850 nm wavelength range. Afterwards, the
270 relationship between $(\alpha \cdot d)^2$ and the photon energy was plotted, assuming that ITO thin
271 films show a direct band gap character [32]. The band gap can be calculated by
272 extrapolating the straight line of the relation between $(\alpha \cdot d)^2$ and photon energy ($h\nu$). The
273 results, portrayed in Fig. 8, show that the change in the energy gap for the ITO thin films
274 is quite small which is in agreement with the overall optical behavior of the three films
275 discussed above. Accordingly, it can be concluded that MwPA combined with RTP does
276 not have a noticeable effect on the optical properties of the ITO thin films, as a
277 consequence of small variations of the index of refraction, having its origin in limited
278 variations of the chemical properties of the thin films. A similar behavior has been
279 previously reported (see, for instance, references [10], [33]).

280

281 **3.5 Electrical properties**

282

283 The electrical behavior of the ITO thin films was determined using the two-point probe
284 method [34]. From the electrical (current-voltage) measurements a linear relationship is
285 found in all cases, as portrayed in Fig. 9, leading to an ohmic electrical behavior.
286 Accordingly, the experimental data were fitted to a linear regression, from which the
287 values of the resistance of every ITO thin film were extracted using Ohm's law [35]. The

288 experimental results show that the annealing process has a notable effect on the resistivity
289 of the ITO thin films, as shown in Table 1. In particular, MwPA processing at 0.1 mbar
290 for 2 minutes cause a remarkable reduction in the resistivity of the ITO thin films,
291 although from an optical point of view the thin film remains opaque. However, applying
292 RTP in oxygen atmosphere at 550°C results in thin films with a large degree of
293 transparency (Fig. 5) and good conductivity. This is probably due to the increased
294 crystalline structure created by the annealing process, as determined by XRD analysis
295 (sec. 3.3), given that grain boundaries in polycrystalline and amorphous materials tend to
296 decrease the electrical conductivity of any given material [36]. In fact, grain boundaries
297 are considered two-dimensional defects in the crystal structure. From Table 1, it is worth
298 mentioning that microwave processing has a very large impact on lowering the resistivity
299 of the deposited films. However, as determined in the previous section, the thin films are
300 still opaque. As such, we resorted to using both methods combined together, i.e.,
301 MwPA+RTP and RTP+MwPA. As a result of the combined post-formation processes,
302 the ITO thin films show a quite large optical transparency and high conductivity.
303 However the best results are obtained for the films treated by RTP first which has a
304 resistivity equal $4.3 \times 10^{-4} \Omega \cdot \text{cm}$ as indicated in Table 1. Table 2 presents a comparison
305 between the previously reported electrical and optical properties of ITO thin films
306 prepared by different methods and for the best combined process of the present work.
307 According to Table 2, the optical and electrical properties for (MwPA+RTP) sample is
308 better than the previously obtained results by e-beam evaporation due to the combination
309 of the thermal processing with MwPA.

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321 Table 2: Summary of the optical and electrical properties of ITO thin films previously
 322 manufactured by different techniques compared with the best results obtained in this work.

Manufacturing Technique	Average transmission ($\lambda= 300-800$ nm)	Resistivity $\times 10^{-4}$ ($\Omega.cm$)	Reference
RF magnetron sputtering	87.5	5.3	[11]
DC Magnetron Sputtering	79	5	[13]
Reactive low voltage ion plating	84	4	[17]
e-beam evaporator	75	5.8	[18]
RF magnetron sputtering	88	< 10	[32]
e-beam evaporator	85	4.3	Present work

323

324

4. DISCUSSION

325

326

327 Table 3 provides a summary of the key properties of the ITO thin films before and after
 328 being subjected to the different post-formation treatments. The experimental results
 329 indicate that the as-deposited and MwPA ITO thin films are opaque, while the rest of the
 330 thin films (processed by RTP, MwPA+RTP, and RTP+MwPA) are transparent.
 331 Additionally, the MwPA ITO thin films show the highest conductivity.

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343 Table 3: Summary of the optical, structural, and electrical properties of the ITO thin films.

ITO thin films	Optical properties					Crystal structure	Resistivity $\pm 0.5 \times 10^{-4}$ ($\Omega \cdot \text{cm}$)
	T%	R%	(<i>n</i>)	(<i>k</i>)	(α)		
As deposited	Opaque thin films	The highest R% average over all the wavelength range	Annealing process causes an increase of (<i>n</i>) for all the treated thin films	Annealing process causes a decrease of (<i>k</i>) for all the treated thin films	High for the opaque films	Amorphous with one broad peak	48.7
MwPA		Low R% at shorter wavelengths, but increases toward the infrared region				Crystalline and the preferential oriented crystal plane is (222)	2.0
RTP	Highly Transparent Thin films	Low R% average over all the wavelength range			Low for the transparent films	16.5	
MwPA+RTP						15.1	
RTP+MwPA			4.3				

344

345 In particular, the as-deposited ITO thin films are opaque and their electrical conduction
 346 properties are inferior to those of the processed thin films. The higher resistivity of ITO
 347 films is generally attributed to the low carrier concentration, which is directly related to
 348 the oxygen content [37]. This way, an oxygen vacancy gives rise to shallow donor states
 349 just below the conduction band by releasing two electrons. Also, one-electron or impurity
 350 states are formed below the In 5s or 5p conduction band by substitution of In^{3+} by Sn^{4+} .
 351 The improvement in the degree of crystallinity causes an increase in the concentration of
 352 electrically active sites, which would increase the carrier concentration [38], [39].

353

354 Microwave energy generates thermal heating associated with atomic vibration and/or
 355 dipole rotation in the thin films [40]. This technique has unique features, including
 356 volumetric heating, i.e., the capability of transferring energy directly to the interior of the
 357 thin film enabling the annealing process to be carried out at low temperature rapidly, and
 358 short processing times [41]. Microwave annealing processes were used before as dopant
 359 activation in silicon and for the recrystallization of amorphous silicon films [42]. As
 360 indicated in Table 3, the MwPA-treated ITO thin films are opaque and their electrical

361 conduction properties are very good. The experimental results show that microwave
362 processing does not change the opaque nature of the as-deposited ITO thin films, probably
363 because oxygen deficiencies still remain. It must be here remembered that microwave
364 processing is carried out in argon. As such, from the optical point of view, the MwPA-
365 processed films show large absorption coefficients. However, the ITO thin films become
366 more conductive. We attribute this increase in the electrical conductivity to the doped tin
367 atoms. The activated dopants, Sn^{+4} ions in our case, would be responsible for substituting
368 In^{+3} to form donor levels in the energy band gap, which leads to an increase in the number
369 of charge carriers. The interaction between the argon gas atoms and the ITO thin film
370 atoms increase the atomic vibrations and, as a result, more ionized Sn^{+4} ions are activated
371 and the generated film transferred from the amorphous state to be crystalline with larger
372 grain size (as determined by FESEM and XRD analysis) than the as-deposited films.

373

374 From Table 3 it is observed that RTP-processed ITO thin films are transparent and good
375 conductors. Moreover, the electrical properties of RTP and (MwPA+RTP)-processed ITO
376 thin films are comparable, although quite different from those of (RTP+MwPA)-
377 processed thin films. In our opinion, thermal annealing under oxygen atmosphere
378 gradually re-oxidizes the In and Sn particles and oxygen vacancies are thus formed again,
379 which increases the conductivity of the thin films. Besides, the degree of crystallinity of
380 the ITO thin films increases with particles of large grain size. The previously mentioned
381 effects also contribute to the larger transparency of the thin films, with low absorption
382 coefficients.

383

384 Combining the two post-formation processes, i.e., RTP and MwPA, results in transparent
385 and low-resistivity thin films due to oxygen vacancies generated by the thermal annealing
386 process in oxygen. (RTP+MwPA)-processed thin films show higher conductivity than
387 (MwPA+RTP)-processed thin films. This behavior might occur because microwave
388 annealing after rapid thermal processing activates more Sn^{+4} ions due to atomic
389 vibrations. In this case, Sn^{+4} ions would substitute In^{+3} to form donor levels in the band
390 gap, thus leading to an increased number of charge carriers as discussed before. However,
391 oxygen vacancies would not be affected by microwave annealing, so the ITO thin film
392 would preserve its large transparency, as indicated in the optical properties section.

393

394 Finally, it is worth pointing out that the (MwPA+RTP)-processed thin films have almost
395 the same optical and electrical properties as RTP-processed thin films, although showing
396 lower conductivity than (RTP+MwPA)-processed thin films. We hypothesize that rapid
397 thermal processing after microwave annealing results in the oxidation of Sn and In metal
398 particles. Accordingly, the observed decrease in conductivity would be of the same order
399 of magnitude as that of RTP-treated thin films. In the three cases, the crystalline structure
400 would not be affected.

401

402

5. CONCLUSIONS

403

404 The effect of four different annealing processes on the morphological, chemical, optical,
405 and electrical properties of ITO thin films grown by electron beam evaporation has been
406 analyzed in this work.

407

408 From the point of view of their morphology, it was found that MwPA processing results
409 in increased the grain size from 2 nm for as deposited ITO thin films to around 18 nm
410 while, at the same time, the electrical resistivity decreases from 48.7×10^{-4} to 2.0×10^{-4}
411 $\Omega \cdot \text{cm}$. However, the thin films are still opaque due to oxygen deficiency. Furthermore, it
412 was found that combining MwPA with RTP leads to a notable improvement in the optical
413 transparency of the ITO thin films in addition to their crystalline structure and a good
414 electrical conductivity (in the order of $10^3 \text{ S} \cdot \text{cm}^{-1}$). Additionally, it was determined that
415 the chemical composition of the ITO thin films is slightly affected by the different post-
416 formation treatments (MwPA, RTP, and the two processes combined). Besides, ITO thin
417 films treated by MwPA after RTP show the highest conductivity with very good degree
418 of transparency, which makes this process the most appropriate for the fabrication of ITO
419 thin films for applications in optoelectronics as transparent electrodes, including
420 electroluminescent devices, flat panel displays, and solar cells.

421

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428

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7. FIGURES CAPTIONS

538

539 Figure 1: Cross section FESEM images of the different ITO films: (1) as-formed, (2)
540 MwPA, (3) RTP, (4) MwPA+RTP and (5) RTP+MwPA processed thin films.

541 Figure 2: EDX analysis of the ITO thin films: (1) as-formed, (2) MwPA, (3) RTP, (4)
542 MwPA+RTP, (5) RTP+MwPA processed thin films and (6) variation of the
543 wt.% of the main elements (In and Sn).

544 Figure 3: XRD patterns for as-deposited and annealed ITO thin films. The Miller indices
545 of the different planes are indicated.

546 Figure 4: Transmittance and reflectance spectra of the as-deposited ITO thin film and
547 upon MwPA processing. Note: both T spectra are superposed.

548 Figure 5: Transmittance and reflectance spectra of the ITO thin films after RTP,
549 MwPA+RTP and RTP+MwPA processing.

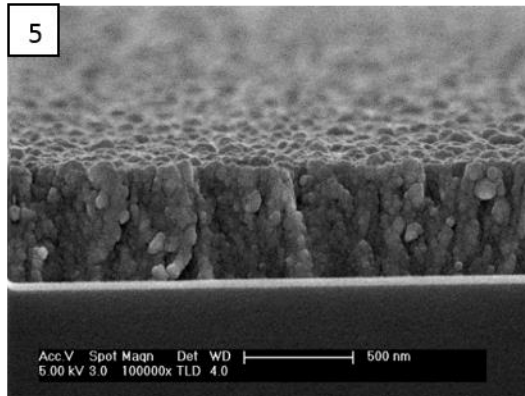
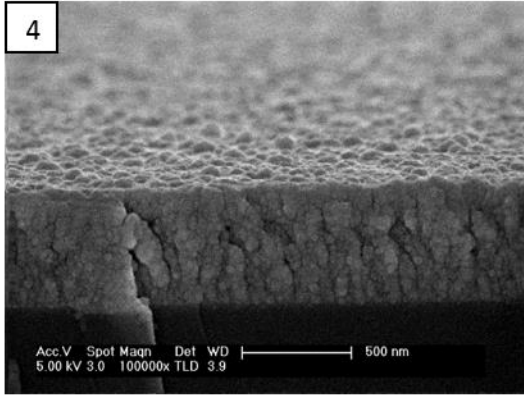
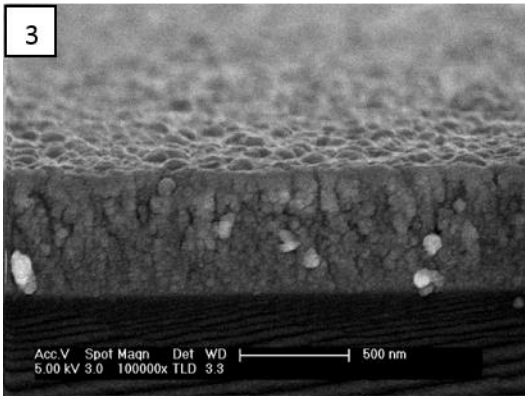
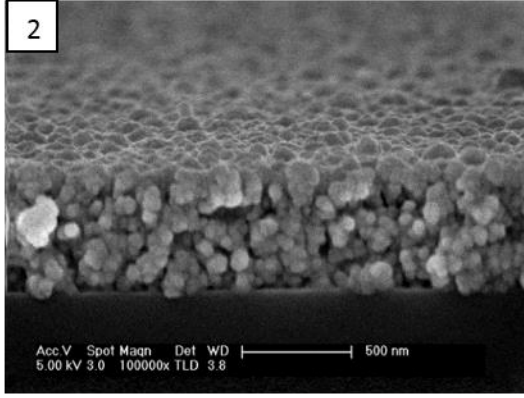
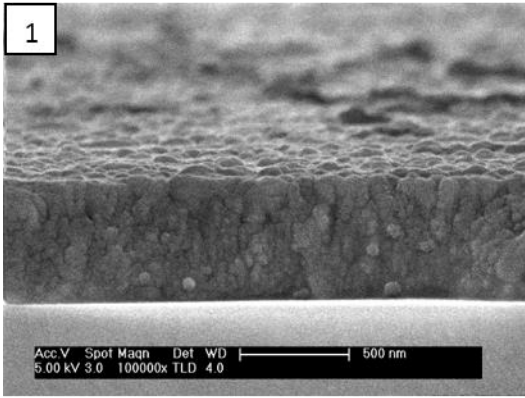
550 Figure 6: Variation of the refractive index with wavelength for the as-deposited and
551 processed ITO thin films.

552 Figure 7: Variation of the extinction coefficient (k) with wavelength for the as-deposited
553 and processed ITO thin films.

554 Figure 8: Tauc's plots used to estimate the values of the energy gap of the transparent thin
555 films.

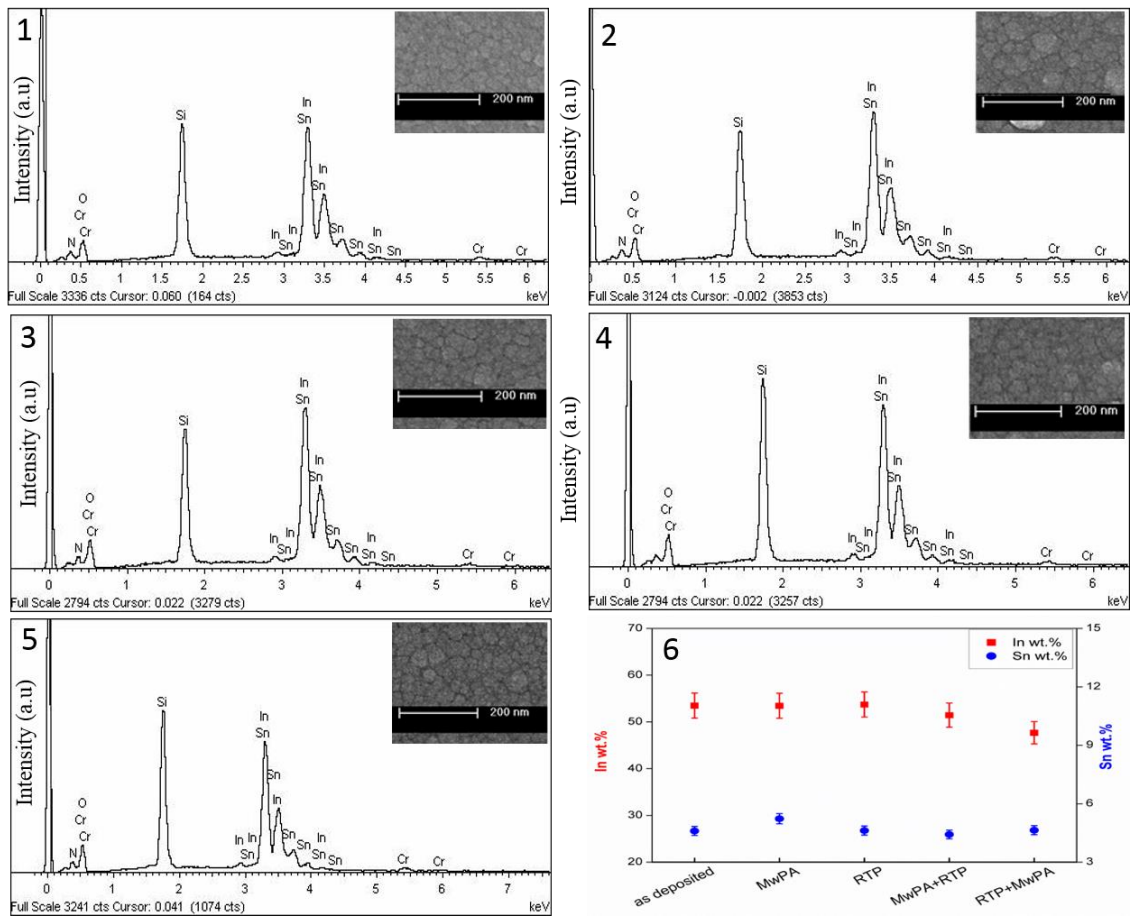
556 Figure 9: Current-voltage (I - V) curve of the different ITO films with different treatments.

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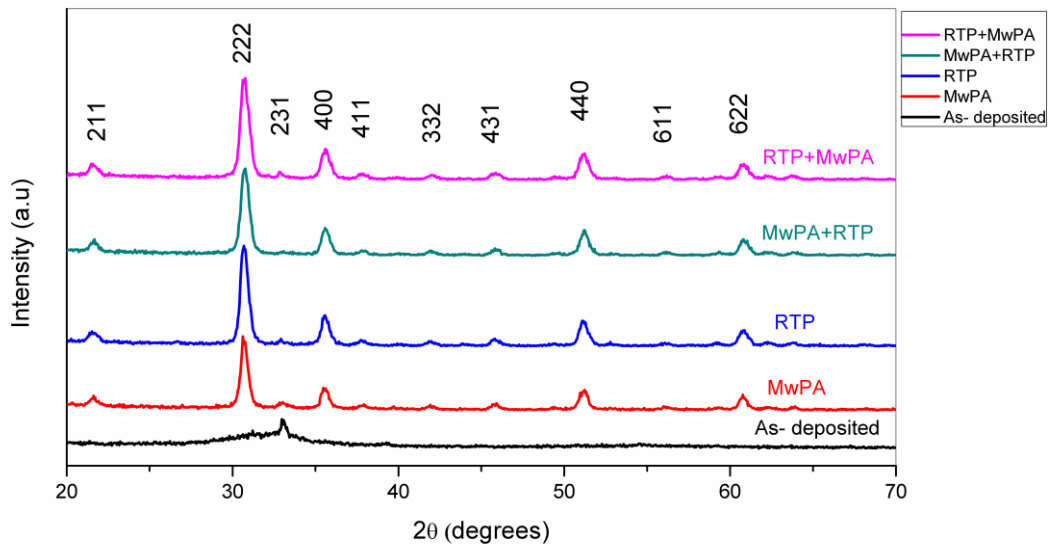


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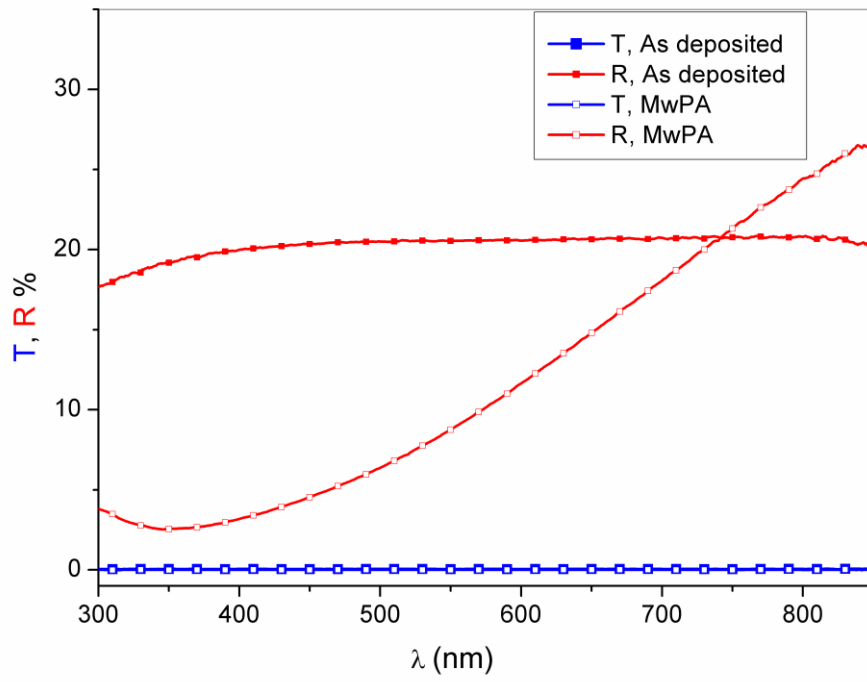
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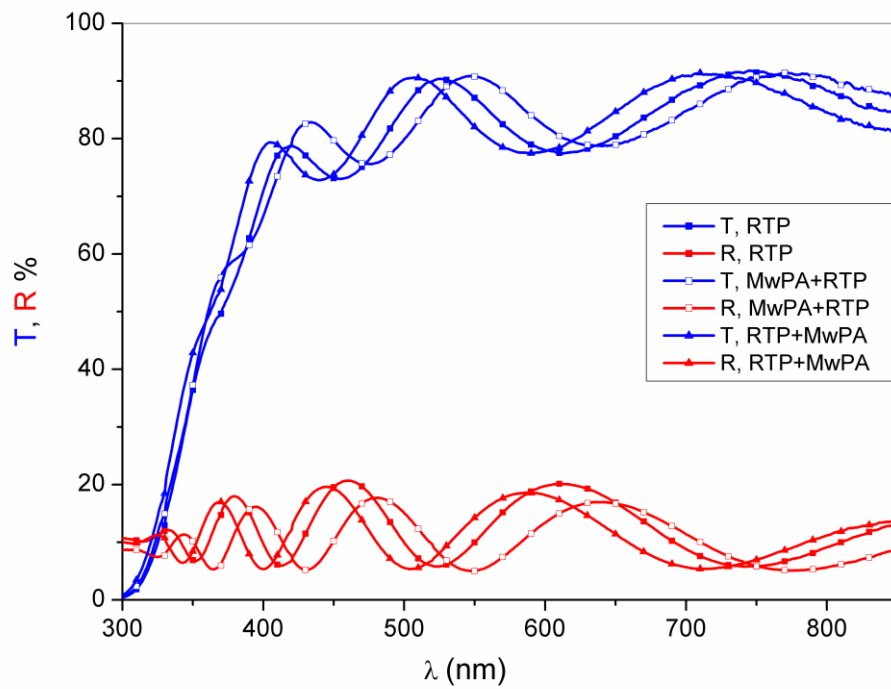


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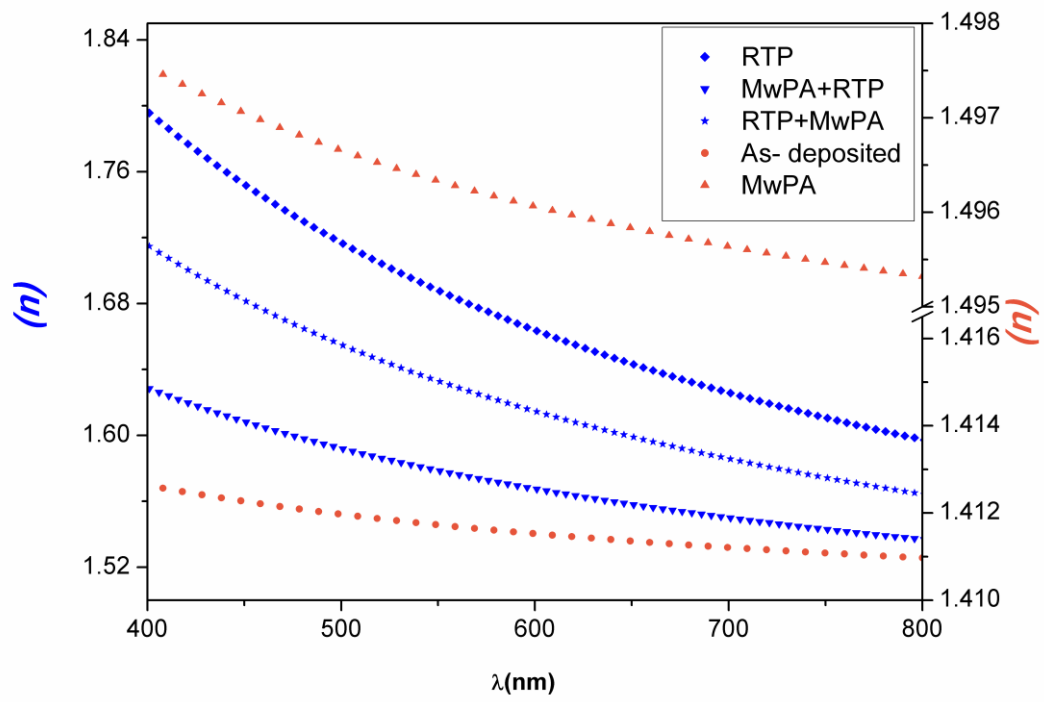
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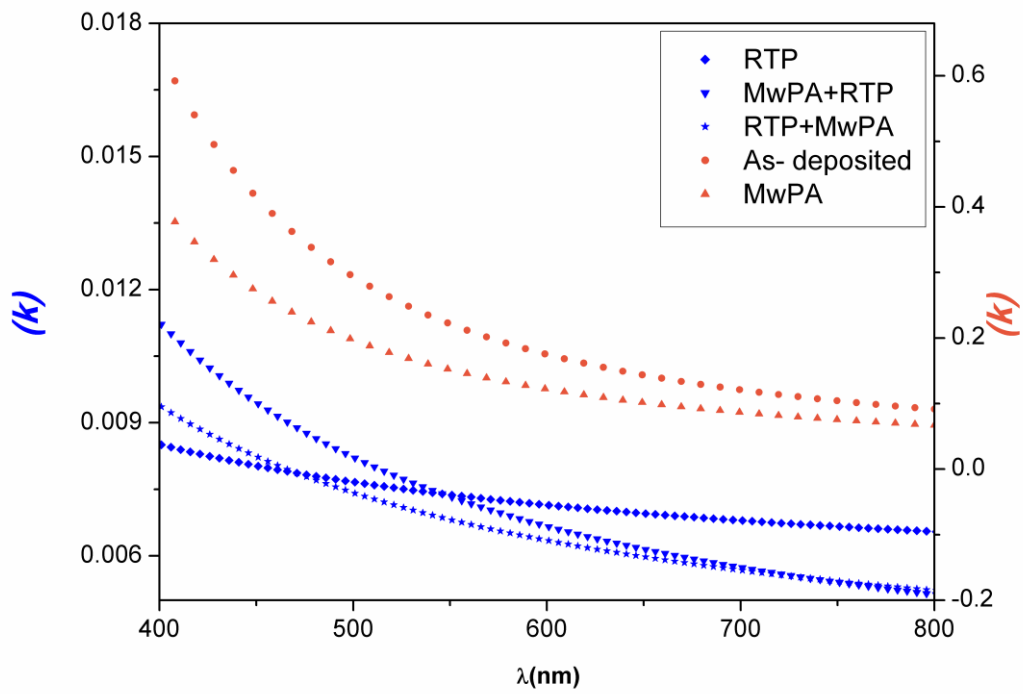
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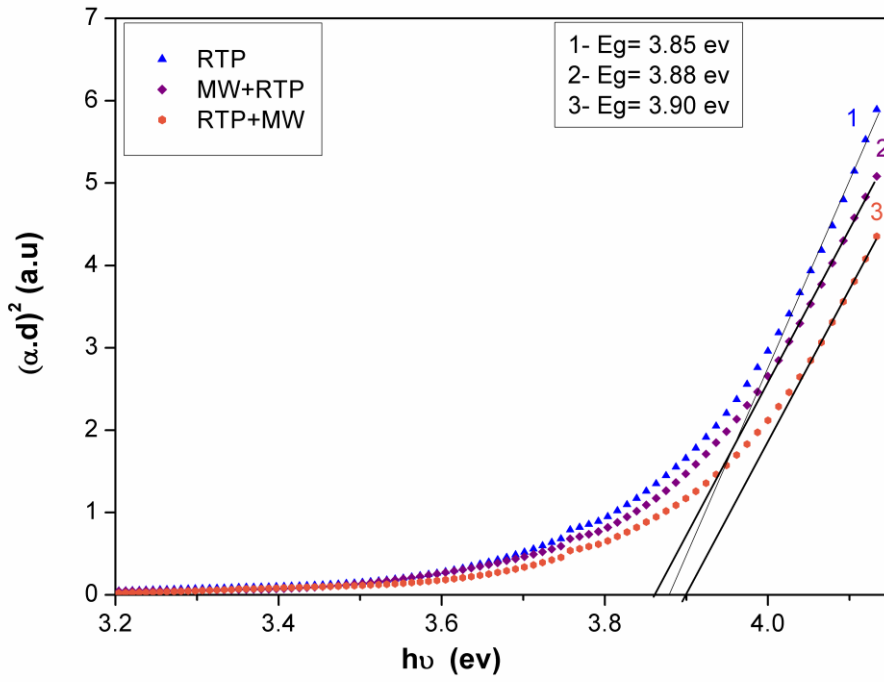
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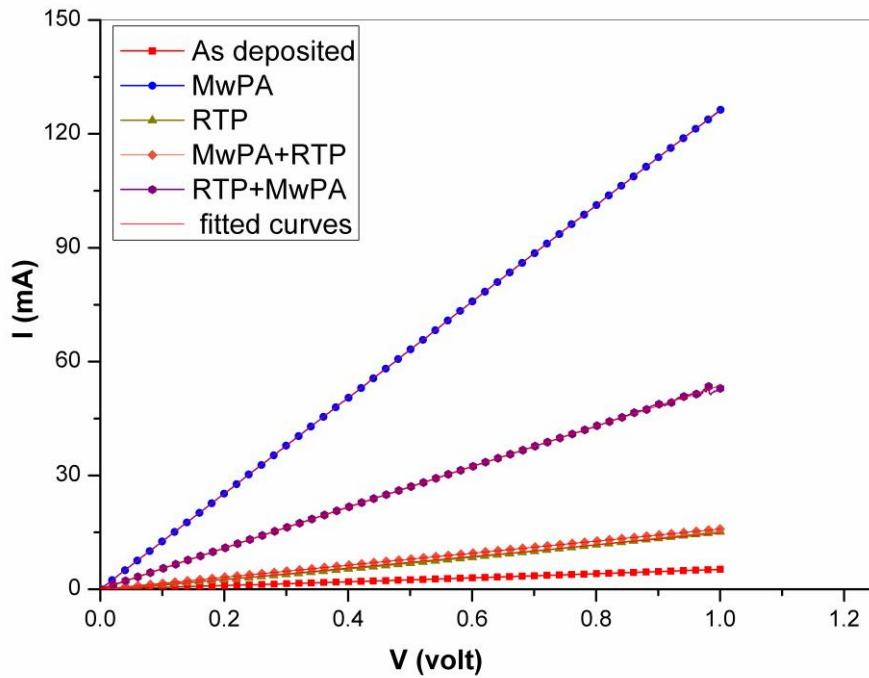
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