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PAPER

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# High performance of solution-processed SnO<sub>2</sub> thin-film transistors by promotion of photo-exposure time-dependent carrier transport during the pre-annealing stage

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## Abstract

We fabricate high-performance solution-processed SnO<sub>2</sub> thin-film transistors (TFTs) exhibiting improved carrier transport features by exposing the ultraviolet/ozone (UV/O<sub>3</sub>) on the SnO<sub>2</sub> film during the pre-annealing stage. The SnO<sub>2</sub> layer is treated with different UV/O<sub>3</sub>-exposure times from 0 to 60 minutes before the post-annealing step. As UV/O<sub>3</sub>-exposure time increases from 0 to 30 minutes, the M-O-M (M, metal; and O, oxygen) network, mass density, and oxygen vacancies of films are enhanced. In contrast, the M-O-M network and mass density decrease, while the oxygen vacancies rather increase when the UV/O<sub>3</sub>-exposure time reaches 60 minutes beyond 30 minutes. The SnO<sub>2</sub> (Sn<sup>4+</sup>) phase, thickness, and surface morphology of SnO<sub>2</sub> films are not considerably changed regardless of UV/O<sub>3</sub>-exposure time. When the UV/O<sub>3</sub>-exposure time is 30 minutes, devices demonstrate superior field-effect mobility (10.1 cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup>) at approximately two times higher than the TFT without UV/O<sub>3</sub>-exposure. Furthermore, the SnO<sub>2</sub> TFT with UV/O<sub>3</sub>-exposure time for 30 minutes shows improved subthreshold-swing characteristics and a high on/off current ratio. These devices are adequate for use in high-resolution active-matrix LCDs or OLED displays that demand a high field-effect mobility (>10 cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup>) and on/off ratio (>10<sup>6</sup>).

Keywords: SnO<sub>2</sub>, photo-exposure time, solution-process, thin-film transistor, high-performance

(Some figures may appear in colour only in the online journal)

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## 1. Introduction

Metal oxide semiconductor-based thin-film transistors (TFTs) have become important as fundamental devices for next-generation displays because they exhibit not only outstanding electron mobility but also high transparency in the visible light regime compared with conventional silicon (Si)-based TFTs [1–4]. Among the different metal oxides, binary oxide semiconductors including indium(III) oxide ( $\text{In}_2\text{O}_3$ ), zinc oxide ( $\text{ZnO}$ ), and tin(IV) oxide ( $\text{SnO}_2$ ) show higher electrical conductivity than the other insulating ionic compounds [5–7]. Many investigators have recently turned to Sn-based oxide TFT because indium (In) is toxic, a rare earth element, and costly [8, 9]. Furthermore, the  $\text{SnO}_2$  semiconductor has many advantages such as a wide optical band gap, crystallization at relatively low temperatures, and remarkable charge transport properties via heavy-metal cations with  $(n-1)d^{10}ns^0$  ( $n \geq 5$ ) electronic configurations, leading to the impressive electrical performance of  $\text{SnO}_2$  TFTs [10–12]. On the other hand, oxide semiconducting materials can be formed into thin-film by both a vacuum process and a solution process [13–16]. From the viewpoint of vacuum-free deposition, the solution process is cost-effective, simple, and applicable to spin-coating, inkjet printing, and meniscus-guided coating, suitable for large-scale production [17–20]. The solution-process inevitably requires a pre-annealing step to evaporate residual solvent on coated films before post-annealing with a relatively high-temperature to develop the active layer [21, 22]. The pre-annealing temperature leads to an endothermal reaction related to the precursor dissociation and subsequent hydrolysis reaction that generates M-OH bonding (M, metal; O, oxygen; and H, hydrogen) which could be converted to M-O-M network bonding through dehydration and alloying reactions during the post-annealing step [21–23]. In this regard, researching pre-annealing treatment is important for improving the electrical performance of oxide TFTs. However, many studies have focused on post-annealing treatments such as high-pressure annealing, ambient gas control, and water vapor annealing [24–26].

In this study, we fabricated high-performance TFTs by controlling ultraviolet/ozone (UV/ $\text{O}_3$ )-exposure time on solution-processed  $\text{SnO}_2$  films during the pre-annealing stage. The change of molecular structure, crystallinity, and morphological characteristics of solution-processed  $\text{SnO}_2$  thin-films with different UV/ $\text{O}_3$ -exposure times from 0 to 60 minutes was systematically investigated. We observed the enhanced physical- and chemical characteristics of  $\text{SnO}_2$  films that show increased mass density, M-O-M network, and oxygen vacancies when the UV/ $\text{O}_3$ -exposure time is 30 minutes. In contrast, mass density and M-O-M network decreased, while the oxygen vacancies rather excessively increased when UV/ $\text{O}_3$ -exposure time reached 60 minutes beyond 30 minutes. The crystalline size, thickness, and surface roughness of  $\text{SnO}_2$  films were similar regardless of UV/ $\text{O}_3$  treatment. The  $\text{SnO}_2$  TFT with UV/ $\text{O}_3$  treated for 30 minutes demonstrated superior electrical properties with the highest field-effect mobility of  $10.1 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  and on/off current ratio of  $1.2 \times 10^6$ ,

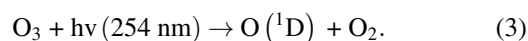
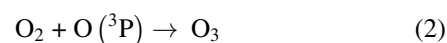
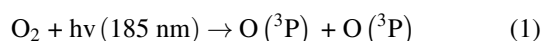
adequate for use in high-resolution active-matrix LCDs or OLED displays [27].

## 2. Methods

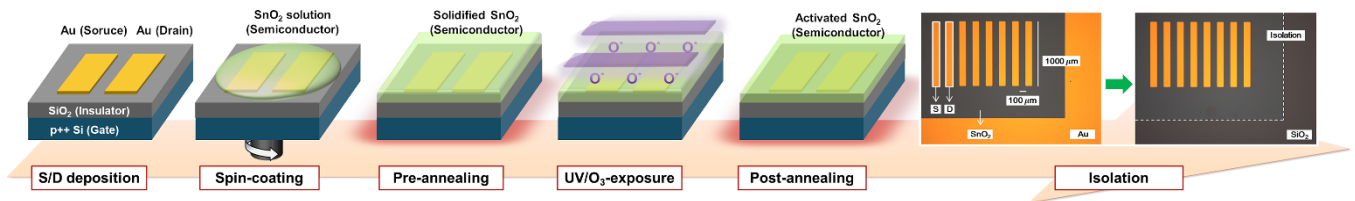
### 2.1. Fabrication process

A schematic for the fabrication process of a coplanar bottom-gate  $\text{SnO}_2$  TFT with UV/ $\text{O}_3$  treatment is depicted in figure 1. A heavily boron-doped p-type Si wafer and thermally grown 100 nm  $\text{SiO}_2$  were used as the gate electrode and gate insulator, respectively. For the bottom contact structure, 50 nm thick Au electrodes were deposited on  $\text{SiO}_2$  using e-beam evaporation and then patterned by the lift-off method to design an active channel layer with a length of 1 000  $\mu\text{m}$  and width of 100  $\mu\text{m}$ . The Si/ $\text{SiO}_2$  substrate was cleaned by UV/ $\text{O}_3$  for 60 minutes before spin-coating. The solution for the  $\text{SnO}_2$  active materials was prepared by dissolving 0.025 M of tin(II) chloride dihydrate ( $\text{SnCl}_2 \cdot 2\text{H}_2\text{O}$ ) in ethanol. The prepared solution was spin-coated at 3000 rpm for 50 s onto a Si/ $\text{SiO}_2$  substrate followed by pre-annealing at 150  $^\circ\text{C}$  for 10 minutes to evaporate the residual solvent. In a previous study,  $\text{SnO}_2$  films coated with ethanol-based chloride precursor solution dried at 150  $^\circ\text{C}$  demonstrated suitable environmental stability with high densification [28].

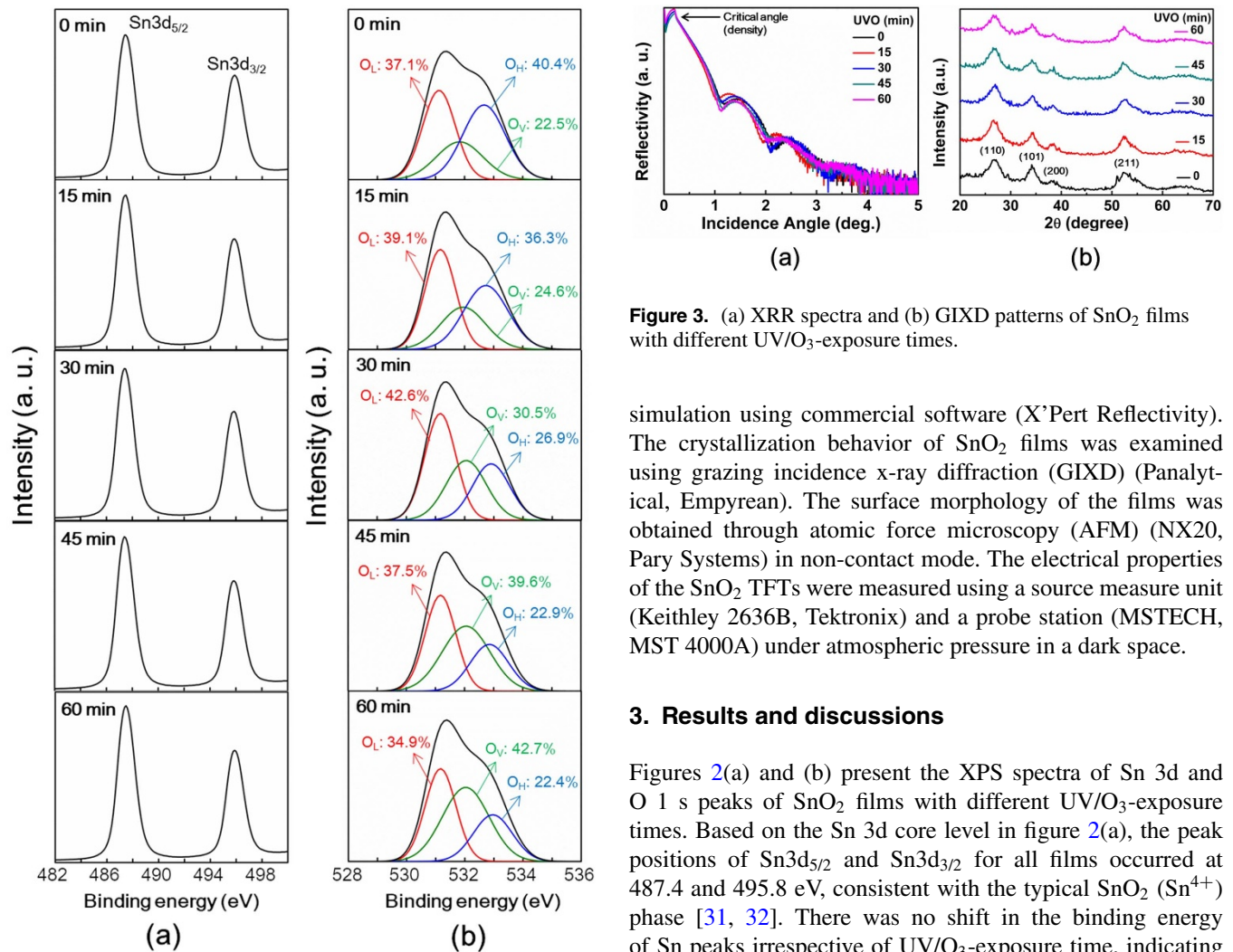
UV irradiation ( $25 \text{ mW cm}^{-2}$ ) was conducted with a low-pressure mercury lamp with two types of light sources (Jaesung engineering, UVC-30). One had a wavelength of 184.9 nm and an energy of  $647 \text{ kJ mol}^{-1}$ , and the other had a wavelength of 253.7 nm and an energy of  $472 \text{ kJ mol}^{-1}$ . These two light source energies are larger than the bonding energy of the contaminant organic and hydrogen compounds such as C-C ( $348 \text{ kJ mol}^{-1}$ ), C-O ( $352 \text{ kJ mol}^{-1}$ ), C-H ( $413 \text{ kJ mol}^{-1}$ ), and O-H ( $463 \text{ kJ mol}^{-1}$ ). Moreover, these light sources continuously produce  $\text{O}_3$  and oxygen free radicals ( $\text{O}^3\text{P}$  and  $\text{O}^1\text{D}$ ) through the following reactions [29, 30]:



As presented in Equations (1) and (3), Ozone ( $\text{O}_3$ ) and excited singlet atomic oxygen ( $\text{O}^1\text{D}$ ) are strong oxidants and easily eliminate organic contaminants as volatile byproduct molecules such as  $\text{CO}_2$ ,  $\text{H}_2\text{O}$ , and  $\text{O}_2$  [29]. The UV/ $\text{O}_3$ -exposure time was adjusted at 0, 15, 30, 45, and 60 minutes to explore the effect of UV/ $\text{O}_3$ -exposure on  $\text{SnO}_2$  film and obtain the high electrical performance of a  $\text{SnO}_2$  TFT. After the pre-annealing stage with UV/ $\text{O}_3$ -exposure, post-annealing was performed at 450  $^\circ\text{C}$  for 1 h in a tube furnace with ambient air to remove organic residues, activate metal oxide film, and assist crystallization of  $\text{SnO}_2$ . The coated films were mechanically eliminated in advance before measurement of the electrical performance in the  $\text{SnO}_2$  TFT for complete



**Figure 1.** A schematic for the fabrication process of the coplanar-bottom gate UV/O<sub>3</sub>-exposed SnO<sub>2</sub> TFT.



**Figure 2.** XPS spectra of (a) Sn 3d and (b) O 1 s state of SnO<sub>2</sub> films with different UV/O<sub>3</sub>-exposure times.

isolation to minimize gate leakage current and fringing effect. The optical images before and after the isolation of the device are shown in figure 1.

## 2.2. Analysis method

x-ray photoelectron spectroscopy (XPS) (ThermoFisher Scientific, NEXSA) was conducted using a monochromatic Cu K $\alpha$  (1.5418 Å) light source to observe the surface electronic states and chemical compositions of SnO<sub>2</sub> films. The mass density and thickness of the films were investigated with x-ray reflectivity (XRR) (Panalytical, Empyrean) analysis and

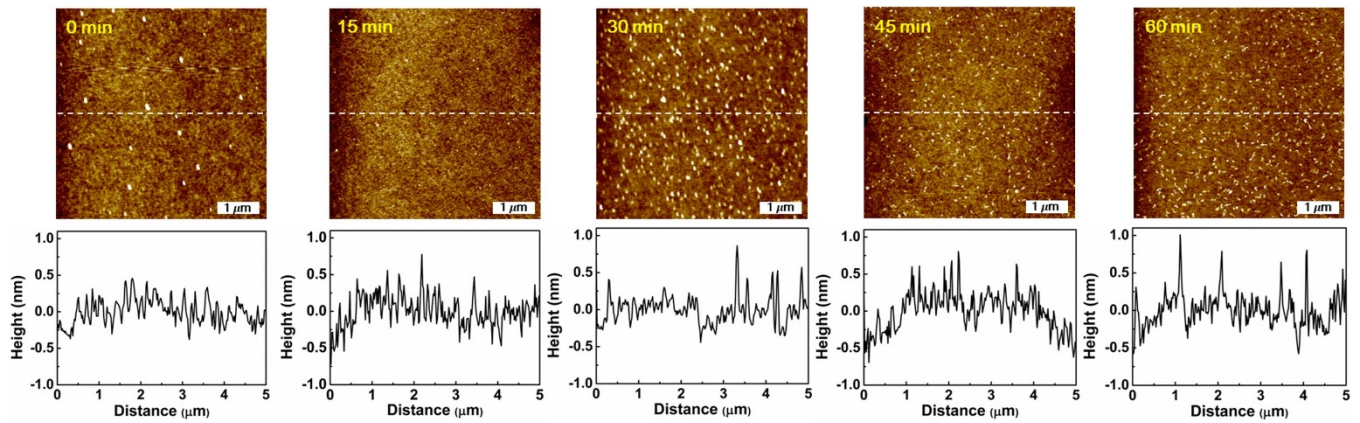
**Figure 3.** (a) XRR spectra and (b) GIXD patterns of SnO<sub>2</sub> films with different UV/O<sub>3</sub>-exposure times.

simulation using commercial software (X'Pert Reflectivity). The crystallization behavior of SnO<sub>2</sub> films was examined using grazing incidence x-ray diffraction (GIXD) (Panalytical, Empyrean). The surface morphology of the films was obtained through atomic force microscopy (AFM) (NX20, Pary Systems) in non-contact mode. The electrical properties of the SnO<sub>2</sub> TFTs were measured using a source measure unit (Keithley 2636B, Tektronix) and a probe station (MSTECH, MST 4000A) under atmospheric pressure in a dark space.

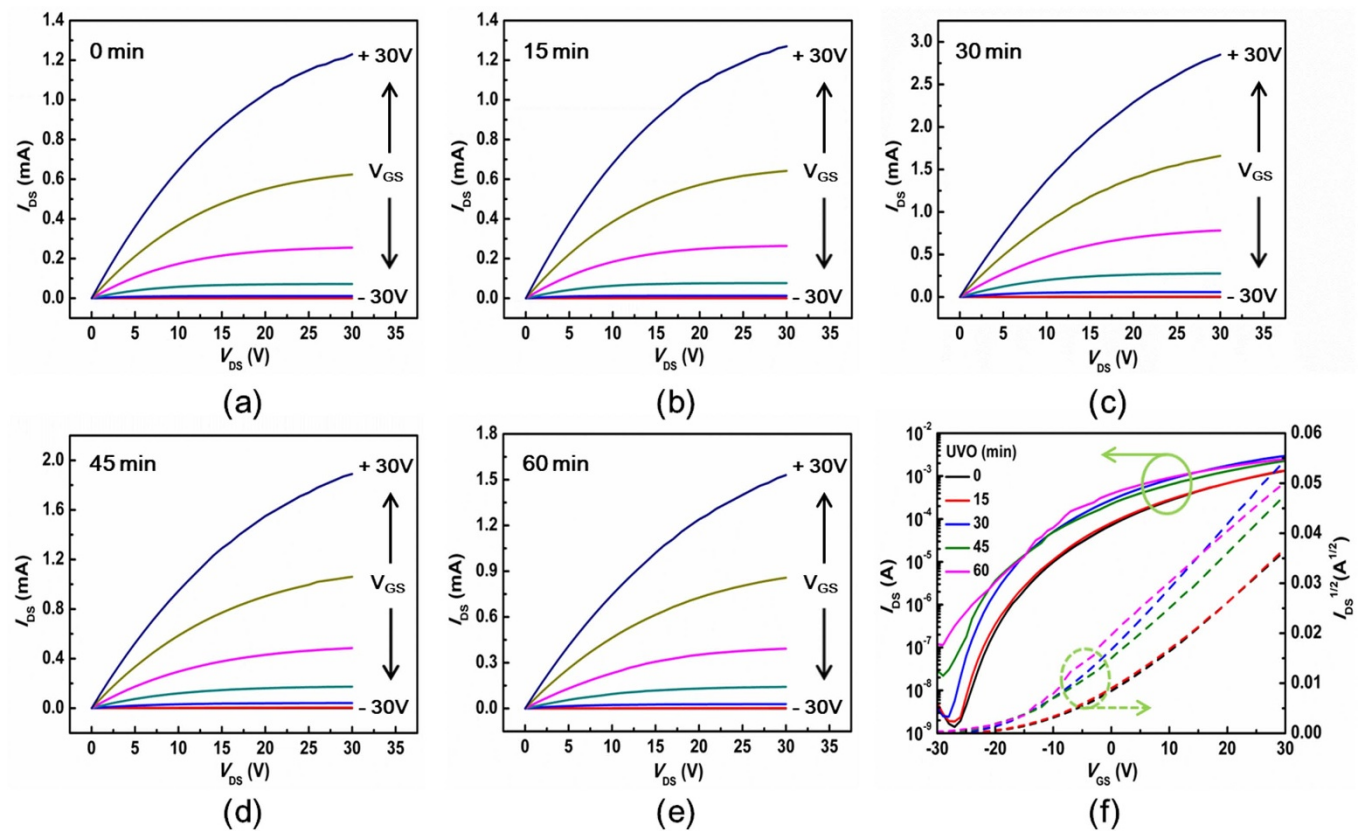
## 3. Results and discussions

Figures 2(a) and (b) present the XPS spectra of Sn 3d and O 1 s peaks of SnO<sub>2</sub> films with different UV/O<sub>3</sub>-exposure times. Based on the Sn 3d core level in figure 2(a), the peak positions of Sn3d<sub>5/2</sub> and Sn3d<sub>3/2</sub> for all films occurred at 487.4 and 495.8 eV, consistent with the typical SnO<sub>2</sub> (Sn<sup>4+</sup>) phase [31, 32]. There was no shift in the binding energy of Sn peaks irrespective of UV/O<sub>3</sub>-exposure time, indicating that UV/O<sub>3</sub>-exposure on SnO<sub>2</sub> films during the pre-annealing stage did not affect the phase transition between SnO<sub>2</sub> and SnO. The O 1 s state is divided into three peaks associated with lattice oxygen (O<sub>L</sub>, denoting the M-O-M network for the electron path), oxygen-vacancies (O<sub>V</sub>, denoting free carrier concentration), and hydroxyl groups (O<sub>H</sub>, denoting the electron trap site) at 531.1, 532.0, and 532.8 eV, using Gaussian fitting as depicted in figure 2(b) [33]. As the UV/O<sub>3</sub>-exposure time increase, the O<sub>H</sub> ratio decreased and the O<sub>V</sub> ratio increased because UV irradiation and the oxidation by both O<sub>3</sub> and oxygen-free radicals contribute to decompose the residual hydrogen compounds and then removed as volatile products. When the UV/O<sub>3</sub>-exposure time was 30 minutes, the O<sub>L</sub> ratio was highest because of improved the M-O-M network formations and reorganization through reduced carbon-based





**Figure 4.** AFM images ( $5 \times 5 \mu\text{m}$ ) and corresponding cross-sectional height profiles of  $\text{SnO}_2$  films with different  $\text{UV}/\text{O}_3$ -exposure times.



**Figure 5.** (a)–(e) Output and (f) transfer characteristics of  $\text{SnO}_2$  TFTs with different  $\text{UV}/\text{O}_3$ -exposure times.

content [16]. However, the  $\text{O}_\text{L}$  ratio was reduced by eliminating too many hydrogen compounds which could be involved in formation of the M-O-M network via condensation reactions when the  $\text{UV}/\text{O}_3$ -exposure time exceeded 30 minutes. Figure 3(a) illustrates the XRR curves of  $\text{SnO}_2$  films with different  $\text{UV}/\text{O}_3$ -exposure times and arrow means the point where the critical angle was determined using X'Pert Reflectivity software. Accordingly, we extracted the critical angle to calculate the mass density of  $\text{SnO}_2$  thin-films using following equation:

**Table 1.** The critical angle, mass density and RMS roughness of the  $\text{SnO}_2$  films with different  $\text{UV}/\text{O}_3$ -exposure times.

$\text{UV}/\text{O}_3$ -exposure time (min)	Critical angle (deg)	Mass density ( $\text{g cm}^{-3}$ )	RMS roughness (nm)
0	0.2417	3.17	0.20
15	0.2420	3.18	0.21
30	0.2613	3.71	0.24
45	0.2680	3.90	0.26
60	0.2424	3.19	0.32

**Table 2.** The electrical properties of the SnO<sub>2</sub> TFTs and their standard deviations with different UV/O<sub>3</sub>-exposure times.

UV/O <sub>3</sub> -exposure time (min)	Field-effect mobility (cm <sup>2</sup> V <sup>-1</sup> s <sup>-1</sup> )		On/off ratio		Subthreshold swing (V/decade)	
	Mean	Std. Dev.	Mean	Std. Dev.	Mean	Std. Dev.
0	5.95	0.43	$8.3 \times 10^5$	$1.2 \times 10^5$	2.53	0.20
15	6.31	0.59	$9.4 \times 10^5$	$2.3 \times 10^5$	2.13	0.21
30	9.81	0.57	$1.1 \times 10^6$	$2.2 \times 10^5$	1.98	0.24
45	6.92	0.64	$1.4 \times 10^5$	$3.5 \times 10^4$	3.55	0.29
60	5.46	0.84	$4.7 \times 10^4$	$1.3 \times 10^4$	3.73	0.45

**Table 3.** Performance comparison of the various solution-processed SnO<sub>2</sub> TFTs.

Ref.	Gate insulator	Capacitance per unit area (nF cm <sup>-2</sup> )	Annealing conditions	Channel length/width (μm)	$\mu_{sat}$ (cm <sup>2</sup> V <sup>-1</sup> s <sup>-1</sup> )	On/off $I_{DS}$ ratio
This work	SiO <sub>2</sub>	34.5	450 °C for 1 h	100/1000	10.1	10 <sup>6</sup>
[38]	SiO <sub>2</sub>	11.5	450 for 4 h	100/600	0.37	10 <sup>6</sup>
[39]	SiO <sub>2</sub>	11.5	450 for 4 h	100/600	0.19	10 <sup>3</sup>
[12]	SiO <sub>2</sub>	31.4	500 for 1 h	5/110	0.23	10 <sup>6</sup>
[40]	SiO <sub>2</sub>	34.5	600 for 4 h	100/1000	10.87	10 <sup>7</sup>
[41]	Al <sub>2</sub> O <sub>3</sub>	225	450 for 30 min	60/1400	96.4	10 <sup>6</sup>

$$\theta_c = \lambda \sqrt{\rho_e r_e / \pi} \quad (4)$$

$$\rho_m = \frac{\rho_e A}{N_A Z} \quad (5)$$

where  $\theta_c$  is the critical angle,  $\lambda$  is the wavelength of x-ray source (1.5418 Å for Cu K $\alpha$ ),  $\rho_e$  is the electron density,  $r_e$  is the classical radius of the electron ( $2.818 \times 10^{-13}$  cm),  $\rho_m$  is the mass density, A is the atomic weight (150.71 g mol<sup>-1</sup> for SnO<sub>2</sub>),  $N_A$  is Avogadro's number ( $6.023 \times 10^{23}$  mol<sup>-1</sup>), and Z is the atomic number (66 for SnO<sub>2</sub>) [34, 35]. While increasing the UV/O<sub>3</sub>-exposure time from 0 to 45 minutes, the mass density of SnO<sub>2</sub> films increased from 3.17 to 3.90 g cm<sup>-3</sup> owing to eliminating organic residue. However, the mass density of SnO<sub>2</sub> films decreased to 3.19 g cm<sup>-3</sup> when the UV/O<sub>3</sub>-exposure time was 60 minutes because the reduced M-O-M network by hydrogen compounds are excessively decomposed, as mentioned in the XPS analysis. Regardless of UV/O<sub>3</sub>-exposure time, the thickness of all SnO<sub>2</sub> films was approximately  $4.53 \text{ nm} \pm 3.6 \text{ Å}$ , calculated with the periodicity of the fringes using XRR simulation. These values are similar to the thickness analyzed by transmission electron microscopy (TEM) in a previous study [11]. We conducted GIXD analysis to investigate the crystallinity of SnO<sub>2</sub> films with different UV/O<sub>3</sub>-exposure times, as depicted in figure 3(b). All diffraction peaks well-matched a tetragonal rutile SnO<sub>2</sub> pattern (JCPDS 41-1445) and had almost the same shape irrespective of UV/O<sub>3</sub>-exposure time. The crystallite size of the films was obtained using the Scherrer formula; all films had a similar crystal size of approximately  $7 \pm 0.8 \text{ Å}$ . Figure 4 illustrates the AFM images and corresponding height profile of SnO<sub>2</sub> films with different UV/O<sub>3</sub>-exposure times. The root-mean-square (RMS) roughness increased from 0.20

to 0.32 nm with increasing UV/O<sub>3</sub>-exposure time up to 60 minutes. Nevertheless, this small surface damage did not affect the electrical properties of the SnO<sub>2</sub> TFT since values of RMS roughness differed only slightly in the angstrom-range and the TFTs are fabricated with bottom-contact structure. The values of critical angle, mass density, and RMS roughness of SnO<sub>2</sub> films with different UV/O<sub>3</sub>-exposure times were summarized in table 1.

Figures 5(a)–(e) exhibit the output characteristics of SnO<sub>2</sub> TFTs with different UV/O<sub>3</sub>-exposure times ( $I_{DS}$ – $V_{DS}$  curves at  $V_{GS}$  from –30 to +30 V with an increment of 10 V). All devices exhibited conventional n-type semiconductor behavior and a normally-turned-on state regardless of UV/O<sub>3</sub>-exposure time. Changes in the transfer characteristics ( $I_{DS}$ – $V_{GS}$ ) of SnO<sub>2</sub> TFT were observed with respect to UV/O<sub>3</sub>-exposure time, as depicted in figure 5(f). All the TFTs were operated in the saturation regime by applying a constant  $V_{DS}$  of +30 V. As UV/O<sub>3</sub>-exposure time increased, the off-current increased and turn-on voltage ( $V_{ON}$ ) shifted slightly negatively from –26 to –28 V as free carrier concentration increased, associated with O<sub>v</sub>. Furthermore, the subthreshold swing characteristics (SS) improved from 2.44 (0 minutes) to 1.83 V/decade (30 minutes) due to reduced organic-based content, whereas SS deteriorated to 4.31 V/decade (60 minutes) since an excessively increased O<sub>v</sub> ratio leads to many oxygen deficiency defects and a high off-current [36, 37]. The field-effect mobility was estimated from the following conventional MOS transistor saturation mobility equation:

$$\mu_{sat} = \frac{2L}{WC_i} \left( \frac{\partial \sqrt{I_{sat}}}{\partial V_{GS}} \right)^2 \quad (6)$$

where  $C_i$  is the gate insulator capacitance per unit area and L and W are the channel length and width. While increasing UV/O<sub>3</sub>-exposure time from 0 to 30 minutes, the extracted

field-effect mobility increased from 6.27 to 10.1  $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$  because of the enhanced M-O-M network and mass density of  $\text{SnO}_2$  films, leading to the highest on-current (on/off ratio of  $1.2 \times 10^6$ ). In contrast, the field-effect mobility decreased to 4.31  $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$  owing to the reduced M-O-M network and high oxygen deficiency defects. The electrical characteristics of the  $\text{SnO}_2$  TFT for different UV/ $\text{O}_3$ -exposure times are summarized in table 2 with both means and standard deviations deduced from 20 devices for each condition. Table 3 shows the performance comparison of the various solution-processed  $\text{SnO}_2$  TFTs with the different fabrication processes. The device of this work exhibits higher field-effect mobility than other studies with a fabrication process analogous to ours (same gate insulator and similar post-annealing temperature). In addition, there is similar field-effect mobility with ours in spite of high post-annealing temperature for a long time. If UV/ $\text{O}_3$ -exposure is applied to other  $\text{SnO}_2$  TFTs with high-k gate insulator or high post-annealing temperature, the electrical performance would be improved as in this work.

#### 4. Conclusion

High-performance solution-processed  $\text{SnO}_2$  TFTs were fabricated by the controlling of UV/ $\text{O}_3$ -exposure on films during the pre-annealing stage. We identified the optimal UV/ $\text{O}_3$ -exposure time for suitable electrical characteristics of  $\text{SnO}_2$  films. When the UV/ $\text{O}_3$ -exposure time was 30 minutes, the  $\text{SnO}_2$  TFTs exhibited the highest electrical performance compared to TFTs without UV/ $\text{O}_3$ -exposure, with field-effect mobility of 10.1  $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$  and on/off ratio of  $1.2 \times 10^6$  resulting from the enhanced M-O-M network, mass density, and oxygen vacancies of films. In contrast, the  $\text{SnO}_2$  TFTs treated with UV/ $\text{O}_3$  exposure time for 60 minutes show inferior field-effect mobility of 6.15  $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$  and on/off ratio of  $2.2 \times 10^4$ , which is lower than TFTs without UV/ $\text{O}_3$ -exposure, because M-O-M network and mass density decreased and oxygen vacancies were excessively increased. Furthermore, significant changes were not observed in the  $\text{SnO}_2$  ( $\text{Sn}^{4+}$ ) phase, thickness, or surface morphology of  $\text{SnO}_2$  films irrespective of UV/ $\text{O}_3$ -exposure time. The  $\text{SnO}_2$  TFT exposed to UV/ $\text{O}_3$  for 30 minutes during the pre-annealing stage demonstrates superior electrical performance, adequate for use in high-resolution active-matrix LCDs or OLED displays that demand a high field-effect mobility ( $>10 \text{ cm}^2 \text{V}^{-1} \text{s}^{-1}$ ) and on/off ratio ( $>10^6$ ).

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