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# The influence of annealing atmosphere on sputtered indium oxide thin-film transistors

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## Abstract

Indium oxide (In<sub>2</sub>O<sub>3</sub>) thin films sputtered at room temperature were annealed under different atmospheres and examined for thin-film transistor (TFT) active channel applications. The annealing process was performed in a rapid thermal annealing system at 350 °C under O<sub>2</sub>, Ar, forming gas (FG, 96% N<sub>2</sub>/4% H<sub>2</sub>), and N<sub>2</sub>. It was found that the annealed In<sub>2</sub>O<sub>3</sub> TFTs exhibited high field-effect mobility ( $\mu_{FE} > 40 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ), high on/off current ratio ( $I_{on/off} \sim 10^8$ ), and controlled threshold voltage ( $V_{TH}$ ) for the enhancement- and depletion-mode operations. Note that the annealing atmosphere has a significant effect on the electrical performance of the In<sub>2</sub>O<sub>3</sub> TFTs by inducing changes in oxygen-related species, particularly oxygen vacancies ( $V_O$ ) and hydroxyl/carbonate species (O–H/C–O). For the O<sub>2</sub>-, Ar-, FG-, and N<sub>2</sub>-annealed TFTs,  $\mu_{FE}$  was in increasing order accompanied by a negative shift in  $V_{TH}$ , which is a result attributed to the larger  $V_O$  in the In<sub>2</sub>O<sub>3</sub> thin films. Furthermore, the  $\Delta V_{TH}$  of the FG-, and N<sub>2</sub>-annealed TFTs in a positive bias stress test was greater than that of the O<sub>2</sub>-, Ar-annealed devices, attributing to their lower density of O–H/C–O groups in the In<sub>2</sub>O<sub>3</sub> thin films. Our results suggest that the annealing atmosphere contributes to the internal modifications of the In<sub>2</sub>O<sub>3</sub> structure and in turn altered the electrical characteristics of TFTs. These annealed In<sub>2</sub>O<sub>3</sub> TFTs with high performance are promising candidates for realizing large-area, transparent, and high-resolution displays.

Supplementary material for this article is available [online](#)

Keywords: In<sub>2</sub>O<sub>3</sub>, thin-film transistor, oxide semiconductor, annealing atmosphere, bias stability

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

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## 1. Introduction

Wide-bandgap oxide semiconductors (OSs) are widely investigated as promising channel layers of thin-film transistors (TFTs) in display technology [1, 2], artificial synapses [3], sensors [4], Schottky devices [5] and other applications [6–8]. Among them, indium–gallium–zinc oxide (IGZO) TFTs have become the backplane standard for active-matrix liquid-crystal displays and active-matrix organic light-emitting diode displays because of their reasonable mobility ( $\mu_{FE} > 10 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ), high  $I_{on/off}$  ratio ( $>10^8$ ), extremely low leakage current ( $<10^{-15} \text{ A}$ ), low process temperature ( $<350^\circ \text{C}$ ), high uniformity, and large-area scalability [9–11]. To meet the requirements of ultra-high-resolution and high frame rate next-generation displays, various OS TFTs with higher mobility than IGZO TFTs have been extensively studied for next-generation display applications [12–14]. Among these transistors, In-rich OS TFTs are the most promising candidates because of their excellent high-mobility performance. The In 5s orbital having a large spatial spread and a large overlap provides a facile electron transport path, which, in addition to their low electron effective mass, endows In-rich OS TFTs with high electron mobility [14].

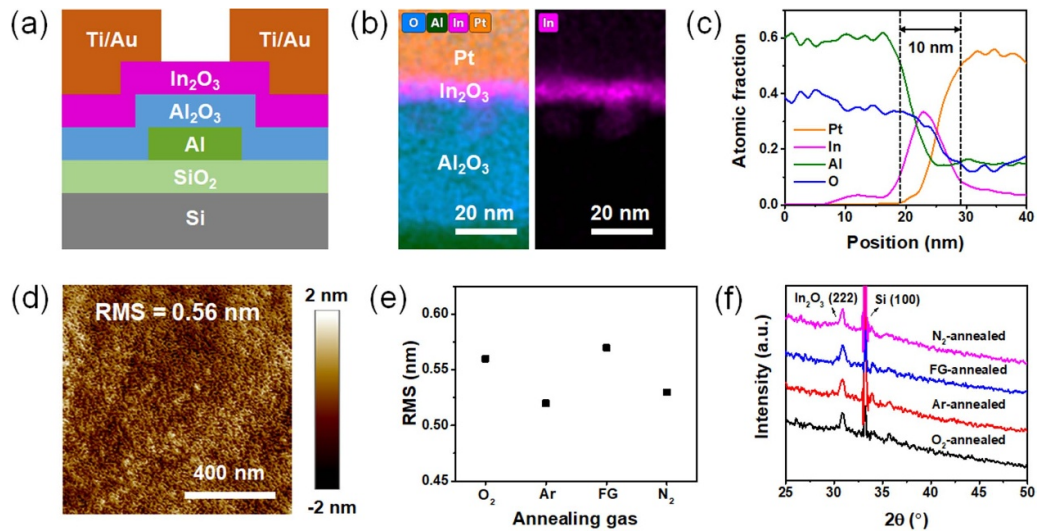
For most OS TFTs, the presence of oxygen-related species, such as oxygen vacancies ( $V_O$ ) or hydroxyl/carbonate species ( $\text{O–H/C–O}$ ), has a considerable impact on their electrical performance [15, 16]. Generally,  $V_O$  readily creates shallow or deep donor states in the oxide channel layer. These shallow donor states supply electrons to the conduction band (CB), leading to an increase in electron concentration. In contrast, the deep donor states in the subgap region act as trap states, degrading device performance [17–19]. In addition, oxygen bonds in  $\text{O–H/C–O}$  groups are considered to generate acceptor-like states near the CB edge and enhance electron trapping during positive bias stress (PBS) applications [20]. Therefore, multiple attempts have been made to modulate oxygen-related species in the active channel layer such as multi-cation doping [21, 22], changing the sputtering gas flow [23], annealing treatment [24], plasma treatment [25], or their combination [26, 27]. One of the effective routines for realizing TFTs with good performance is the deposition of OS channel layers with high resistivity, followed by a post-deposition annealing treatment. Annealing is one of the most commonly used methods for optimizing active channel layers because of its effectiveness in alleviating deep traps formed by ion bombardment or unintended defects during film deposition or the synthesis process [28]. Most previous studies demonstrated that the performance of TFTs under multiple annealing conditions varies depending on the channel material and process conditions chosen [29, 30]. Different annealing conditions, especially the annealing atmosphere, have a significant effect on the performance of OS TFTs. However, most studies on the effects of annealing conditions have focused on IGZO [31], indium–gallium–tin oxide (IGTO) [29], indium–zinc–tin oxide (IZTO) [32], indium–gallium oxide [33], and other In-based multi-cation composition OSs [34, 35]. Few studies have been conducted on the effect of the annealing atmosphere

on the performance of  $\text{In}_2\text{O}_3$  TFTs. Yuan *et al* reported that the sputtered  $\text{In}_2\text{O}_3$  TFTs followed by a post-deposition annealing in vacuum or air showed significantly different mobilities; however, the fundamental mechanism behind the carrier mobility enhancement is unclear [36]. Si *et al* reported that  $\text{In}_2\text{O}_3$  TFTs deposited by atomic layer deposition (ALD) and annealed in  $\text{O}_2$ ,  $\text{H}_2$ , or  $\text{N}_2$  respectively, produced similar results indicating that the annealing atmosphere had no effect on the performance of  $\text{In}_2\text{O}_3$  TFTs [37]. However, ALD-processed and sputtered  $\text{In}_2\text{O}_3$  thin films significantly differ in terms of aspects such as chemical composition and bonding state [38, 39]. Therefore, this conclusion cannot be inferred to be applicable to sputtered  $\text{In}_2\text{O}_3$  TFTs. For OS TFTs, a complete understanding of the effects of the annealing atmospheres on their electrical properties and stability is required. In particular, stability under gate bias stress is important for OS TFT applications. This suggests the need to determine the most suitable annealing atmosphere for  $\text{In}_2\text{O}_3$  thin films to improve the electrical properties of  $\text{In}_2\text{O}_3$  TFTs. In this study, we deposited  $\text{In}_2\text{O}_3$  thin films by sputtering at room temperature and investigated the effect of the annealing atmosphere on the electrical characteristics and bias stability of  $\text{In}_2\text{O}_3$  TFTs.

## 2. Experimental details

The fabrication process of the devices is as follows. First, heavily doped p-type silicon ( $\text{p}^{++}\text{-Si}$ ) substrates with  $2 \mu\text{m}$  thermally grown  $\text{SiO}_2$  were cleaned with acetone, isopropyl alcohol, and deionized water for 10 min each. Next, 30 nm Al was deposited using radio frequency (RF) magnetron sputtering as the gate metal. Then, 30 nm  $\text{Al}_2\text{O}_3$  was grown as the gate dielectric at  $250^\circ \text{C}$  using  $(\text{CH}_3)_3\text{Al}$  (TMA) and  $\text{O}_2$  plasma as Al and O precursors. Next,  $\sim 10 \text{ nm}$   $\text{In}_2\text{O}_3$  films were deposited by RF magnetron sputtering at room temperature as the active channel layer.  $\text{In}_2\text{O}_3$  films with a thickness of  $\sim 40 \text{ nm}$  were then deposited on the  $\text{SiO}_2/\text{Si}$  substrates and sapphire substrates to examine their structural and optical properties. The sputtering power was 100 W, the  $\text{O}_2$  and Ar gas flow ratios were set to 50%, and the working pressure was 10 mTorr. About 5 min pre-sputtering was performed to eliminate the contamination of the  $\text{In}_2\text{O}_3$  target. Subsequently, 20 nm Ti and 100 nm Au were deposited by RF magnetron sputtering as the source and drain metal contacts, respectively. Finally, the  $\text{Al}_2\text{O}_3$  was dry-etched using  $\text{BCl}_3$  as the reactive gas to expose the underlying gate. The bottom gate, channel, and source/drain metals were fabricated by lithography, sputtering and lift-off processes. The channel length and width were 40 and  $240 \mu\text{m}$ , respectively. The annealing process was performed in a rapid thermal annealing system under four different annealing atmospheres of  $\text{O}_2$ , Ar, forming gas (FG) and  $\text{N}_2$ . The annealing temperature and time were fixed at  $350^\circ \text{C}$  and 1 min, respectively.

The surface morphology was characterized using an atomic force microscopy (AFM, Bruker Dimension Icon scanning probe microscope) in tapping mode. Film thickness was jointly determined by transmission electron microscopy (TEM), AFM



**Figure 1.** (a) Schematic of an In<sub>2</sub>O<sub>3</sub> transistor. (b) HAADF-STEM cross-sectional image with EDX elemental mapping of an O<sub>2</sub>-annealed In<sub>2</sub>O<sub>3</sub> transistor, and (c) the corresponding cross-sectional profiles of the elements, confirming that the thickness of In<sub>2</sub>O<sub>3</sub> film is ~10 nm. (d) AFM image of the O<sub>2</sub>-annealed In<sub>2</sub>O<sub>3</sub> thin film. (e) Distribution of surface roughness RMS values extracted from AFM images under different annealing atmospheres. (f) XRD diffraction patterns of all annealed In<sub>2</sub>O<sub>3</sub> thin films.

and ellipsometry. TEM lamella samples were prepared by focused ion beam (FIB) using a FEI Helios G4 scanning electron microscope. Before FIB milling, a platinum (Pt) layer was deposited to protect the sample surface from ion damage. The FEI Titan ST microscope system with an acceleration voltage of 300 kV was equipped with a Super-X energy-dispersive x-ray spectroscopy (EDX) for high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) imaging. Crystal structures were studied using a Bruker D2 PHASER x-ray diffraction (XRD) system with a Cu tube source ( $\lambda = 1.54184 \text{ \AA}$ ) at 30 kV. O 1s spectra were examined by x-ray photoelectron spectroscopy (XPS) was performed in a high vacuum using a Kratos Amicus XPS system equipped with a monochromatic Al K $\alpha$  x-ray source operating at 10 kV. The electrical properties and gate bias stability of the fabricated TFTs were measured using a Keithley 4200 system and a Cascade Summit probe station at room temperature and ambient atmosphere.

### 3. Results and discussion

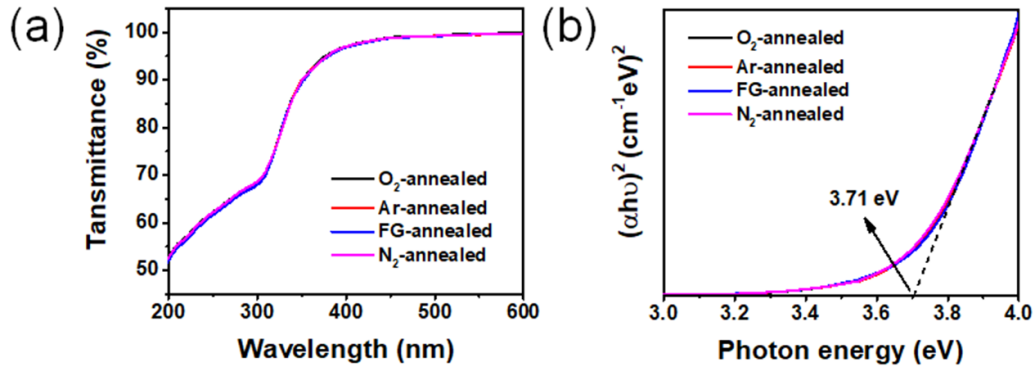
Figure 1(a) shows a schematic of an In<sub>2</sub>O<sub>3</sub> TFT with a bottom-gate-top-contact structure fabricated on a Si substrate. The gate stack is structured with 30 nm Al as the bottom gate, 30 nm Al<sub>2</sub>O<sub>3</sub> as the gate dielectric, ~10 nm In<sub>2</sub>O<sub>3</sub> as the channel layer, and 20/100 nm Ti/Au as the top source/drain contact. Figure 1(b) shows the HAADF-STEM with EDX mapping of the O<sub>2</sub>-annealed In<sub>2</sub>O<sub>3</sub> TFT, highlighting the Al/In/O elements. The top Pt layer was deposited to protect the top portion of the specimen from ion damage during sample preparation. The corresponding elemental profiles shown in figure 1(c) confirm that the thickness of the In<sub>2</sub>O<sub>3</sub> thin film is ~10 nm. The thickness of the In<sub>2</sub>O<sub>3</sub> thin film was also jointly determined

by ellipsometer and AFM measurements. Figure 1(d) shows the surface morphology image of the O<sub>2</sub>-annealed In<sub>2</sub>O<sub>3</sub> thin film, indicating a smooth surface. As shown in figure 1(e), the surface roughness root mean square (RMS) values of the O<sub>2</sub>-, Ar-, FG-, and N<sub>2</sub>-annealed In<sub>2</sub>O<sub>3</sub> films extracted from the corresponding AFM images are 0.56, 0.52, 0.58, and 0.53 nm, respectively, indicating that the surface roughness of the annealed films does not depend on the different annealing atmospheres. Figure 1(f) shows the XRD diffraction pattern of the annealed In<sub>2</sub>O<sub>3</sub> thin films on the SiO<sub>2</sub>/Si substrate. A weak diffraction peak at ~31.8° corresponding to the (222) diffraction pattern was observed for all annealed In<sub>2</sub>O<sub>3</sub> thin films, indicating their crystallinity nature. The presence of a strong diffraction peak at ~33.2° originates from the Si (100) substrate. The crystallinity of the In<sub>2</sub>O<sub>3</sub> thin films was further confirmed by TEM analysis.

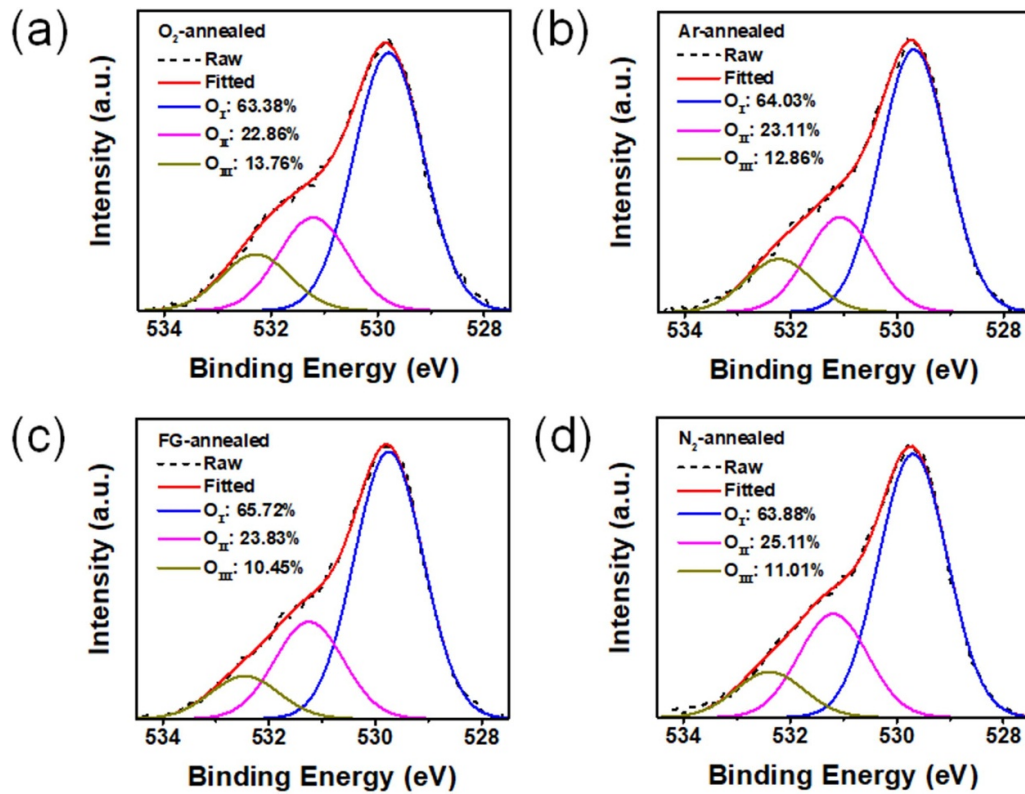
Figure 2(a) shows the UV-Vis transmission spectra of all annealed In<sub>2</sub>O<sub>3</sub> thin films. These films were deposited on a sapphire substrate with a thickness of ~40 nm. The result indicates that the transmittance of the films does not depend on the annealing atmosphere, and all films demonstrate almost full transmittance in the visible region. Figure 2(b) shows the corresponding Tauc plots extracted from the transmission spectra. The optical bandgap ( $E_g$ ) is determined using the Tauc's relation [40, 41]:  $(ahv)^2 = A(hv - E_g)$ , where  $a$  is the absorption coefficient,  $A$  is a constant,  $h$  is the Planck constant, and  $v$  is the frequency.  $E_g$  is extracted by extrapolating the linear part to the energy at  $(ahv)^2 = 0$ . Results indicate that the annealing atmosphere has no effect on the bandgap of the films because all annealed films exhibit the same  $E_g$  value of 3.71 eV.

To investigate the oxygen-related species in the In<sub>2</sub>O<sub>3</sub> thin films annealed under different annealing atmosphere, XPS measurements were performed. As shown in figure 3, the O 1s





**Figure 2.** (a) UV-Vis transmittance spectra, and (b) corresponding extracted Tauc plots of In<sub>2</sub>O<sub>3</sub> thin films.



**Figure 3.** O 1s spectra of In<sub>2</sub>O<sub>3</sub> thin films annealed under (a) O<sub>2</sub>, (b) Ar, (c) FG, and (d) N<sub>2</sub> atmospheres, respectively.

peak was deconvoluted into three peaks centered at 529.7 eV (O<sub>I</sub>), 531.3 eV (O<sub>II</sub>), and 532.4 eV (O<sub>III</sub>), corresponding to the oxygen in the In–O, V<sub>O</sub>, and O–H/C–O [42, 43], respectively. The peak area ratios of O<sub>II</sub>/(O<sub>I</sub> + O<sub>II</sub> + O<sub>III</sub>) were 22.86%, 23.11%, 23.83%, and 25.11% for the In<sub>2</sub>O<sub>3</sub> thin films annealed under O<sub>2</sub>, Ar, FG, and N<sub>2</sub> respectively. These results indicate the presence of a large number of V<sub>O</sub> in the In<sub>2</sub>O<sub>3</sub> thin films. The N<sub>2</sub>- and O<sub>2</sub>-annealed films have the largest and smallest number of V<sub>O</sub>, respectively. Similar results have been reported regarding IGZO TFTs [44, 45]. In addition to V<sub>O</sub>, the O–H/C–O component is an important criterion to determine the quality of the In<sub>2</sub>O<sub>3</sub> thin films, as less O–H/C–O leads to reduced surface scattering and electron charge trapping [29, 46]. The peak area ratios of O<sub>III</sub>/(O<sub>I</sub> + O<sub>II</sub> + O<sub>III</sub>) of the In<sub>2</sub>O<sub>3</sub> thin films annealed under O<sub>2</sub>, Ar, FG, and N<sub>2</sub> were 13.76%,

12.86%, 10.45%, and 11.01%, respectively. To summarize, the O<sub>2</sub>-annealed In<sub>2</sub>O<sub>3</sub> thin film has the smallest V<sub>O</sub> and the largest O–H/C–O component, whereas the Ar-annealed In<sub>2</sub>O<sub>3</sub> thin film exhibits moderate V<sub>O</sub> and O–H/C–O component. Both N<sub>2</sub>- and FG-annealed films exhibit smaller O–H/C–O components, whereas the V<sub>O</sub> component of the N<sub>2</sub>-annealed In<sub>2</sub>O<sub>3</sub> thin film is larger than that of the FG-annealed In<sub>2</sub>O<sub>3</sub> thin film.

Table 1 lists the elemental concentrations of the In<sub>2</sub>O<sub>3</sub> thin films annealed under different atmospheres. The In/O atomic ratios of the annealed In<sub>2</sub>O<sub>3</sub> thin films are in increasing order of the annealing atmosphere of O<sub>2</sub>, Ar, FG, and N<sub>2</sub>, respectively. Moreover, even for the smallest atomic ratio (1:1.39) of the In<sub>2</sub>O<sub>3</sub> thin film annealed in O<sub>2</sub>, it is still larger than that (1:1.5) of stoichiometric In<sub>2</sub>O<sub>3</sub>. This reveals that sputtered In<sub>2</sub>O<sub>3</sub> thin films are typically rich in V<sub>O</sub>.

**Table 1.** Elemental concentrations of In<sub>2</sub>O<sub>3</sub> thin films annealed under different atmospheres.

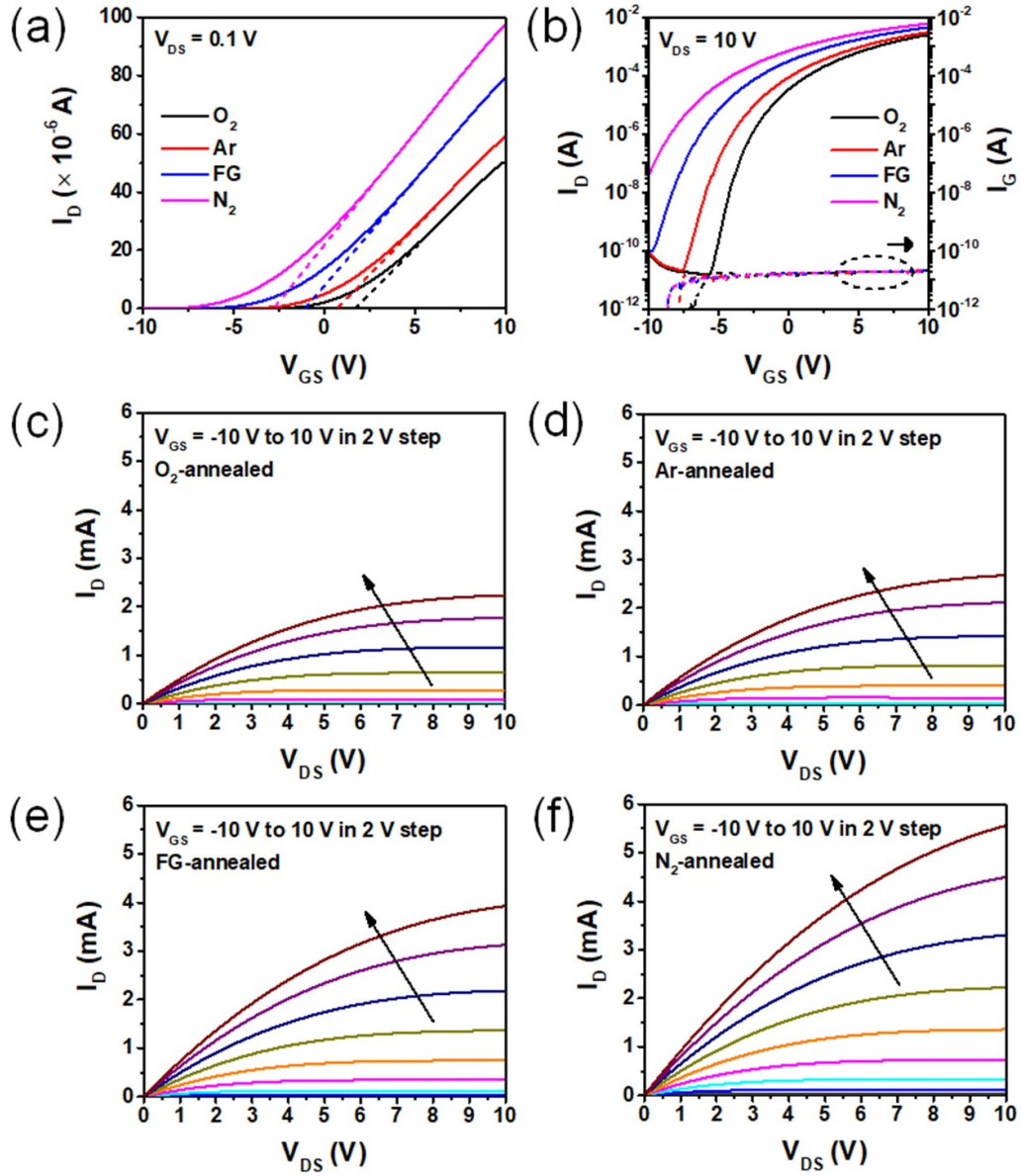
Annealing atmosphere	In	O	Atomic ratio of In:O
O <sub>2</sub>	41.92%	58.08%	1:1.39
Ar	43.61%	56.39%	1:1.29
FG	43.80%	56.20%	1:1.28
N <sub>2</sub>	44.07%	55.93%	1:1.27

Figures 4(a) and (b) shows the representative  $I_D$ - $V_{GS}$  characteristics of In<sub>2</sub>O<sub>3</sub> TFTs annealed under different atmosphere on linear ( $V_{DS} = 0.1$  V) and logarithmic ( $V_{DS} = 10$  V) scales, respectively. Here, the enhancement mode is defined as  $V_{TH} > 0$  V, whereas the depletion mode is the opposite. Using linear extrapolation, the  $V_{TH}$  of the O<sub>2</sub>-, Ar-, FG-, and N<sub>2</sub>-annealed In<sub>2</sub>O<sub>3</sub> TFTs are 1.7, 0.7, -1.1 and -2.8 V, respectively. This result indicates that enhancement- and depletion-mode operation of In<sub>2</sub>O<sub>3</sub> TFTs can be selectively achieved under different post-deposition annealing atmospheres. The  $I_{on/off}$  ratio of the O<sub>2</sub>-, Ar-, and FG-annealed TFT is  $\sim 10^8$ , with the FG-annealed TFT exhibiting a slightly higher  $I_{off}$ . In contrast, the N<sub>2</sub>-annealed In<sub>2</sub>O<sub>3</sub> TFT exhibited undesired switching characteristics that are not completely depleted at a reasonable gate bias. Note that the gate leakage current ( $I_G$ ) for all annealed samples exhibits low values of  $\sim 10^{-11}$  A, which is the minimum current detection limit of our Keithley 4200 system. Figures 4(c)–(f) shows the representative  $I_D$ - $V_{DS}$  characteristics of In<sub>2</sub>O<sub>3</sub> TFTs annealed under different atmosphere. All annealed TFTs exhibit good operation in the n-channel mode with clear pinch-off voltages and saturation drain currents. The linear increase in  $I_D$  in the low drain voltage region shows a good ohmic contact between the channel layer and the source/drain contact.  $I_D$  obtained at  $V_{GS}$  of 10 V is 2.2, 2.7, 3.9, and 5.6 mA in increasing order for the O<sub>2</sub>-, Ar-, FG-, and N<sub>2</sub>-annealed In<sub>2</sub>O<sub>3</sub> TFTs, respectively. The reference as-deposited In<sub>2</sub>O<sub>3</sub> TFT was also measured, and the corresponding  $I_D$ - $V_{GS}$  and  $I_D$ - $V_{DS}$  curves are shown in figure S1. Compared to the annealed samples, the as-deposited In<sub>2</sub>O<sub>3</sub> TFT shows undesirable electrical performance with a larger  $V_{TH}$  (6.8 V) and lower  $I_D$  (0.011 mA) at  $V_{GS}$  of 10 V under the same  $V_{DS}$  sweeping condition. Similar results have been reported for IGZO [47], IZTO [32], and InSnO [48] TFTs. It is known that the OS thin films deposited at room temperature typically have a loose structure that exhibits carrier scattering caused by poor film quality, resulting in few free carriers in the films and low conductivity and mobility. The electrical performance of the films can be greatly improved by promoting oxygen diffusion from the OS channel layer and rearrangement of molecular bonds through post-deposition annealing [32, 47, 48].

Figures 5(a)–(d) summarizes the dependence of  $V_{TH}$ ,  $\mu_{FE}$ ,  $I_D$ , and SS on the annealing atmosphere evaluated from over 20 devices on the same substrate.  $\mu_{FE}$  was determined from the linear transfer characteristics from equation (1),  $\mu_{FE} = Lg_m/(WC_{OX}V_{DS})$ , where  $g_m$  is the transconductance,  $C_{OX}$  is the oxide capacitance of the gate dielectric, and  $W$  and

$L$  are the channel width and channel length, respectively. SS was determined from equation (2):  $SS = (dV_{GS}/d\log I_{DS})_{MAX}$ .  $I_D$  was defined as the  $I_D$  at a fixed  $V_{OV}$  ( $V_{OV} = V_{GS} - V_{TH}$ ) of 8 V for  $V_{DS} = 0.1$  V in the transfer characteristics. All annealed TFTs show narrow parameter distributions in a single batch of devices, which confirms their high uniformity. For TFTs annealed under O<sub>2</sub>, Ar, FG, and N<sub>2</sub>,  $\mu_{FE}$ ,  $I_D$ , and SS are in increasing order, accompanied by a negative shift in  $V_{TH}$ . The observed electrical results are consistent with XPS results shown in figure 3 as we assume that the condition of the In<sub>2</sub>O<sub>3</sub> thin films is correlates with that of oxygen-related species, particularly V<sub>O</sub>. The conducting electrons in the In<sub>2</sub>O<sub>3</sub> thin films are mainly derived from V<sub>O</sub> that act as shallow donor states. Thus, a high number of V<sub>O</sub> indicates high carrier concentration, resulting in low  $V_{TH}$ , a high conductivity and mobility [49, 50]. However, a higher electron concentration promotes the formation of percolation conduction paths in OSs, making it a problem for TFTs to reach complete depletion. The O<sub>2</sub>-annealed In<sub>2</sub>O<sub>3</sub> TFT exhibits lower conductivity and mobility because O<sub>2</sub> fills certain V<sub>O</sub>, resulting in a lower carrier concentration than those of the other films. The N<sub>2</sub>-annealed In<sub>2</sub>O<sub>3</sub> TFT only reached partial depletion because of an excessive carrier concentration, which is mainly attributed to the desorption of oxygen atoms during the N<sub>2</sub> annealing process resulting in a large amount of V<sub>O</sub> in the In<sub>2</sub>O<sub>3</sub> thin film [51]. These results are consistent with previous reports on IGZO and IGTO TFTs [29, 45]. The SS of the N<sub>2</sub>-annealed In<sub>2</sub>O<sub>3</sub> TFT is not provided because  $V_{TH}$  is so negative that SS cannot be meaningfully measured. The higher SS of N<sub>2</sub>- and FG-annealed TFTs than that of the O<sub>2</sub>- and Ar-annealed TFTs indicates their poorer gate control, which is mainly attributed to their higher carrier concentrations.

The dependence of gate bias stability on the annealing atmosphere was investigated with a positive bias value of 10 V and a stress time of 3000 s. During bias stress, the gate was biased at a fixed voltage value of 10 V, while the source and drain were grounded. Bias stability is evaluated by the variation of  $V_{TH}$ , which is determined using the linear extrapolation of the transfer curve at the maximum  $g_m$  point. Figures 6(a)–(d) show the evolution of the transfer characteristics of the In<sub>2</sub>O<sub>3</sub> TFTs annealed under O<sub>2</sub>, Ar, FG, and N<sub>2</sub>, respectively. From these figures, the transfer curves shift almost parallel toward the positive direction as the stress time increases. The long-term rate of  $V_{TH}$  shift is significantly reduced with a clear directional difference, thus suggesting the rebalance of the rates of different degradation mechanisms [52]. From the  $V_{TH}$ -stress time curves in figure 6(e), the corresponding  $\Delta V_{TH}$  of the O<sub>2</sub>-, Ar-, FG-, and N<sub>2</sub>-annealed TFTs are 4.8 V, 3.9 V, 3.3 V and 3.4 V, respectively, at a stress time of 3000 s. This  $\Delta V_{TH}$  difference showed the same trend as the difference of relative area of the O<sub>III</sub> component observed in the XPS analysis. Previous studies on OS thin films and TFTs have demonstrated that that O<sub>III</sub> mainly originates from the oxygen in absorbed hydroxyl and carbonate species (O-H/C-O), which typically generate acceptor-like states near the CB edge and enhance electron trapping during PBS application because of their polar nature in OSs [29]. Therefore, the larger



**Figure 4.** Representative (a) linear-scale and (b) log-scale  $I_D$ - $V_{GS}$  characteristics of In<sub>2</sub>O<sub>3</sub> TFTs prepared under different annealing atmospheres. Representative  $I_D$ - $V_{DS}$  characteristics of (c) O<sub>2</sub>-, (d) Ar-, (e) FG, and (f) N<sub>2</sub>-annealed In<sub>2</sub>O<sub>3</sub> TFTs.

$\Delta V_{TH}$  observed in the PBS tests of the O<sub>2</sub>- and Ar-annealed In<sub>2</sub>O<sub>3</sub> TFTs can be attributed to the higher density of O-H/C-O groups within the channel layer.

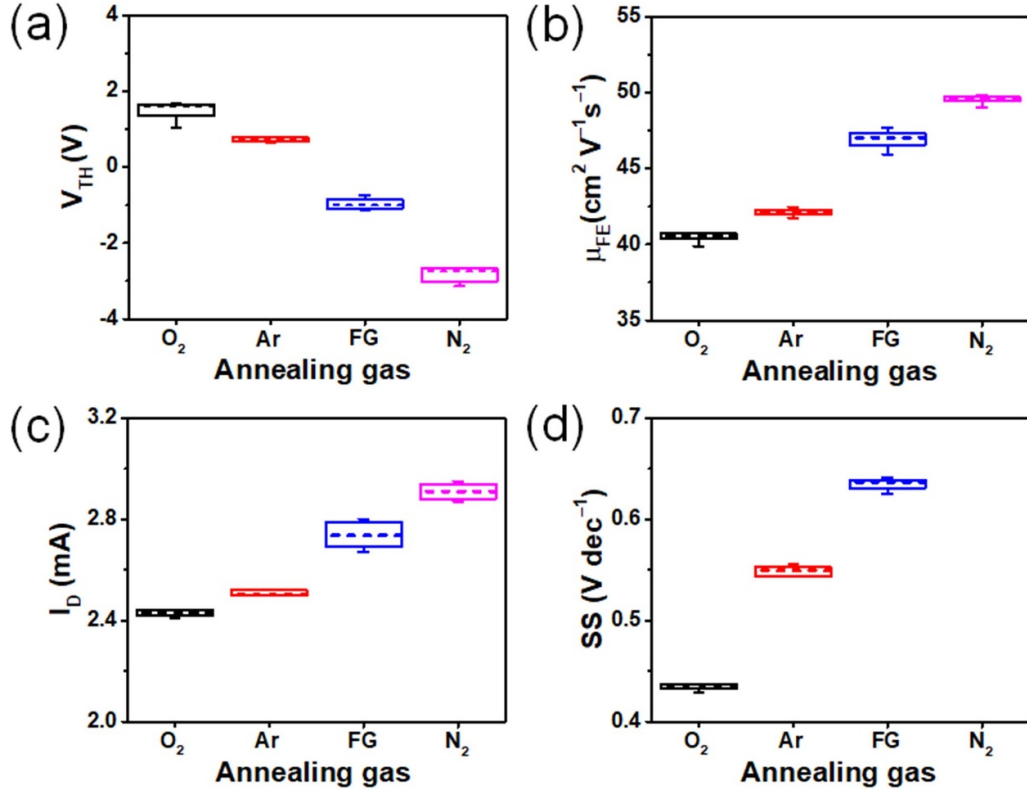
It is known that there are two factors that contribute to  $V_{TH}$  instability: (1) defect creation in the channel and (2) charge trapping in the dielectric and/or at the channel/insulator interface [53]. Defect creation typically results in the persistent degradation of sub-threshold slope and device mobility, whereas charge trapping does not [54]. The main difference between charge trapping at the interface and the injection into the dielectric is the amount of energy needed to remove the injected charge. Releasing the charge injected into a dielectric requires high energy and typically requires thermal annealing or the application of bias [55]. We observed that our devices can automatically relax to the original states within 4 h at room temperature after bias testing. The spontaneous

recovery of  $V_{TH}$  after relaxation without any high energy and the negligible changes in SS shown in figure 6(f) suggest that charge trapping at the channel/insulator interface is the main reason for the instability of the In<sub>2</sub>O<sub>3</sub> TFT PBS condition. The SS of the N<sub>2</sub>-annealed In<sub>2</sub>O<sub>3</sub> TFT is not provided in figure 6(f) because its large negative  $V_{TH}$  so that SS cannot be meaningfully measured.

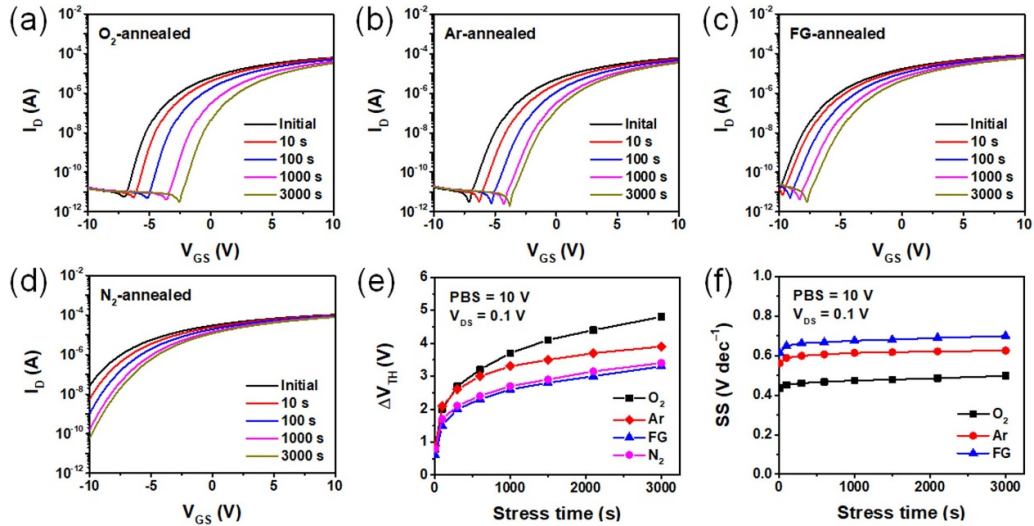
The time-dependent  $V_{TH}$  observed during the PBS tests for the TFTs annealed under different atmospheres can be fitted using the stretched exponential equation:

$$\Delta V_{TH}(t) = \Delta V_{TH0} [1 - \exp[-(t/\tau)^\beta]]$$

where  $\Delta V_{TH0}$  is  $\Delta V_{TH}$  at infinite time,  $t$  is the stress time,  $\beta$  is the stretched-exponential exponent, and  $\tau$  is the characteristic trapping time of the carriers, which correlates



**Figure 5.** Dependence of the (a)  $V_{TH}$ , (b)  $\mu_{FE}$ , (c)  $I_D$ , and (d)  $SS$  of annealed  $In_2O_3$  TFTs under different annealing atmospheres, respectively. The boxes are drawn with 25% of the data as the minimum and 75% as the maximum. The short horizontal lines correspond to the median line of the data.



**Figure 6.** Evolution of transfer characteristics of  $In_2O_3$  TFTs annealed in (a)  $O_2$ , (b)  $Ar$ , (c)  $FG$ , and (d)  $N_2$  under PBS conditions. Variation in (e)  $\Delta V_{TH}$  and (f)  $SS$  as a function of stress time.

with the average effective energy barrier [56, 57]. Table 2 lists the exacted fitting parameters from curve fitting for all TFTs. The  $In_2O_3$  TFTs annealed under different atmospheres have different  $\Delta V_{TH0}$ ,  $\tau$  and  $\beta$ , indicating different device degradation mechanisms, attributed to the generation of undesirable trap centers [52]. Furthermore, certain studies confirmed that the dynamic interaction between the exposed backchannel and the ambient atmosphere affects the  $V_{TH}$  stability of TFTs in

PBS tests. The adsorbed oxygen can capture electrons from the CB, thus resulting in different oxygen species such as  $O^{2-}$  and  $O^-$ . As a result of charge transfer, a depletion layer is formed beneath the oxide surface, thus leading to a positive shift in the  $V_{TH}$  of the transistor. Because our devices use a back-channel structure without a passivation layer covering it, the influence of the ambient atmosphere cannot be excluded [58–60].



**Table 2.** Summary of stretched-exponential fitting parameters for the PBS conditions of In<sub>2</sub>O<sub>3</sub> TFTs under different annealing atmospheres.

Annealing atmosphere	$\Delta V_{TH0}$ (V)	$\tau$ (s)	$\beta$
O <sub>2</sub>	10.97	$1.78 \times 10^4$	0.31
Ar	6.12	$2.79 \times 10^3$	0.26
FG	5.56	$4.60 \times 10^3$	0.30
N <sub>2</sub>	5.67	$4.45 \times 10^3$	0.29

#### 4. Conclusion

In this study, the effects of annealing atmospheres (O<sub>2</sub>, Ar, FG, and N<sub>2</sub>) on the structural and electrical properties of room-temperature sputtered In<sub>2</sub>O<sub>3</sub> thin films as active channel layers were investigated. All annealed In<sub>2</sub>O<sub>3</sub> thin films exhibit polycrystalline properties with high transparency and optical band gap of 3.71 eV. The annealing atmosphere has a significant effect on the electrical properties and bias stability of In<sub>2</sub>O<sub>3</sub> TFTs, which is mainly attributed to significant changes in oxygen-related species, particularly V<sub>O</sub> and O–H/C–O. The measured  $\mu_{FE}$  and SS values of the TFTs annealed under different atmospheres were in the order of O<sub>2</sub> (lowest) to Ar, FG, and N<sub>2</sub> (highest). In the same order, the  $V_{TH}$  of TFTs became increasingly negative. This was mainly attributed to the higher electron concentration caused by the higher V<sub>O</sub> concentration in the In<sub>2</sub>O<sub>3</sub> active channel layer. All annealed In<sub>2</sub>O<sub>3</sub> TFTs exhibit high mobilities ( $>40 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ) and microampere level of  $I_D$ . The O<sub>2</sub>-, Ar-, and FG-annealed In<sub>2</sub>O<sub>3</sub> TFTs exhibit clear transfer characteristics ( $I_{on}/I_{off} \sim 10^8$ ) because of a proper modulation of V<sub>O</sub> in these In<sub>2</sub>O<sub>3</sub> thin films. However, the N<sub>2</sub>-annealed TFT exhibits undesired switching properties that are not completely depleted at a reasonable gate bias because of a high carrier concentration caused by excess V<sub>O</sub>. In addition, the  $\Delta V_{TH}$  values observed in the PBS tests for the FG-, and N<sub>2</sub>-annealed TFTs were smaller than those for the O<sub>2</sub>-, Ar-annealed TFTs, which is attributed to the lower density of O–H/C–O groups in the In<sub>2</sub>O<sub>3</sub> active channel layer. Charge trapping at the channel/insulator interface was considered to be the main factor contributing to PBS instability, and the  $\Delta V_{TH}$  observed during the PBS tests agreed well with the stretched exponential equation. Our study suggests that the annealing of sputtered In<sub>2</sub>O<sub>3</sub> thin films is an effective approach to achieve high-performance TFTs with high mobility and controllable  $V_{TH}$ . Considering the strong influence of the annealing atmosphere on the electrical properties of In<sub>2</sub>O<sub>3</sub> TFTs, careful selection of the annealing atmosphere is necessary.

#### Data availability statements

All data that support the findings of this study are included within the article (and any supplementary files).

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