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Facile Routes To Improve Performance of Solution-Processed Amorphous Metal Oxide Thin Film Transistors by Water Vapor Annealing

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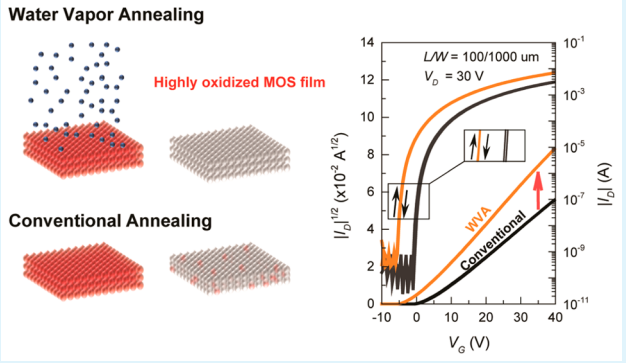
Supporting Information

ABSTRACT: Here, we report on a simple and high-rate oxidization method for producing solution-based compound mixtures of indium zinc oxide (IZO) and indium gallium zinc oxide (IGZO) metal-oxide semiconductors (MOS) for thin-film transistor (TFT) applications. One of the issues for solution-based MOS fabrication is how to sufficiently oxidize the precursor in order to achieve high performance. As the oxidation rate of solution processing is lower than vacuum-based deposition such as sputtering, devices using solution-processed MOS exhibit relatively poorer performance. Therefore, we propose a method to prepare the metal-oxide precursor upon exposure to saturated water vapor in a closed volume for increasing the oxidization efficiency without requiring additional oxidizing agent. We found that the hydroxide rate of the MOS film exposed to water vapor is lower than when unexposed (≤18%). Hence, we successfully fabricated oxide TFTs with high electron mobility (27.9 $cm^2/V{\cdot}s$) and established a rapid process (annealing at 400 °C for 5 min) that is much shorter than the conventional as-deposited long-duration annealing (at 400 °C for 1 h) whose corresponding mobility is even lower (19.2 $cm^2/V{\cdot}s$).



KEYWORDS: *metal oxide semiconductor, indium gallium zinc oxide, indium zinc oxide, thin-film transistor, solution process, water vapor annealing*

1. INTRODUCTION

Amorphous oxide semiconductor (AOS) is one of the rapidly emerging materials for applications in thin-film transistors (TFTs) in order to drive active matrix organic light-emitting diodes (AMOLED).[1−3] The charge transport in AOS film is not sensitive to the film crystallinity since it takes place through the isotropic ns orbital of the metals in the AOS lattice.[4] As a result, TFTs based on AOS are intrinsically suited for flexible electronics owing to the absence of high-temperature annealing that is otherwise needed to obtain the high film crystallinity that is often required for the desired device performance. Recently, flexible and transparent indium gallium zinc oxide (IGZO) TFTs have been demonstrated as sputtered AOS on plastic substrates.[3] Solution-based sol−gel processes to form AOS thin films are also a promising technique that may create a new paradigm of manufacturing large-area,[5] transparent, and flexible devices by using cost-effective graphic-art printing technologies. However, sol−gel processed metal-oxide TFTs still suffer from inferior device performance, reliability, and uniformity as compared to the vacuum-based sputtering counterparts.[6,7] Furthermore, it is difficult to clarify the exact chemical composition and structure of sol−gel processed AOS films by conventional chemical analyses. In addition, in order to obtain reliable device performance, sol−gel processes still necessitate additional pre/postdeposition treatments such as high-temperature thermal annealing,[8] ozone exposure,[9] precursor synthesis,[10] and adding agent.[11] Those methods would be too difficult to realize because they all mainly focus on thorough removal of the undesired reaction byproducts and on the effective formation of metal−oxygen (M−O) bonds inside the as-spun oxide film.

Among all of the above methods, thermal annealing is the most widely used method to improve device characteristics because it is very effective in removing residual solvents,

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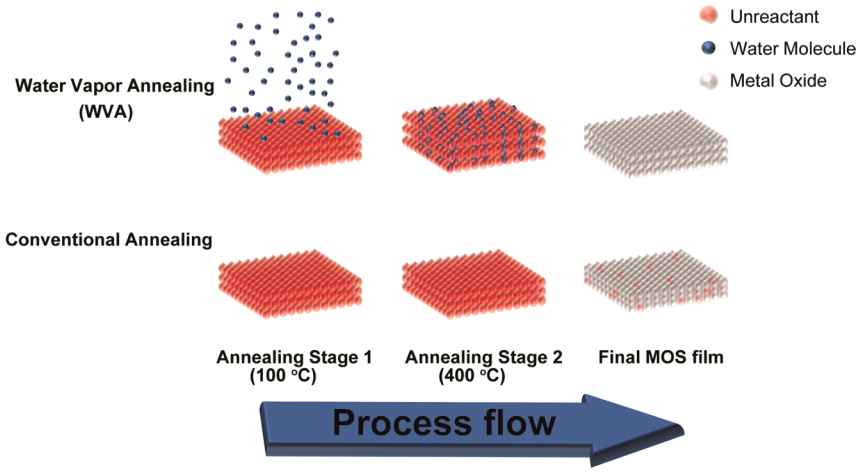

Figure 1. Illustration of WVA and conventional metal-oxide semiconductor film formation processes.

unreacted reactants, and impurities and in formatting M−O bonds from as-spun oxide film. A number of parameters should be considered when seeking to reach an optimal condition, such as annealing temperature and environmental atmosphere. In general, metal-oxide precursors require high-temperature annealing (>300 °C) to be converted into oxidized components. Decreasing the annealing temperature may produce oxide films containing too much hydroxide and poorly organized bonds to the neighboring atoms, which are both detrimental to device performance and stability. Many recent works reported reduced-temperature annealing for flexible device applications but these methods require additional complex processing and/or chemical preparations so that they are very difficult to apply to real industrial manufacturing.[9−11] In this paper, we report on a simple water-vapor annealing method for improving the performance and stability of solution-processed indium zinc oxide (IZO) and IGZO metal-oxide TFTs that is very promising for practical applications.

2. EXPERIMENTAL SECTION

Preparation of Metal Oxide Solution. The IZO and IGZO precursor solutions were prepared using indium(III) nitrate hydrate (99.9% trace metals basis, Sigma−Aldrich), zinc nitrate hydrate (99.999% trace metals basis, Sigma−Aldrich), and gallium(III) nitrate hydrate (crystalline, 99.9% trace metals basis, Sigma−Aldrich). Mother solutions (Zn, In, Ga precursor), 0.1 M in metal precursors, were prepared in 2-methoxyethanol (anhydrous, 99.8%, Sigma−Aldrich) and stirred by magnetic bar for 2 h at room temperature before use. The IZO and IGZO precursors were mixed by measuring the volume ratio from the 0.1 M mother solution. All reagents were used without further purification. The IZO precursor solution composition volume ratio was In/Zn = 1:1, and the IGZO precursor solution composition volume ratio was In/Ga/Zn = 6:1:3. These solutions were stirred by magnetic bar for 2 h at room temperature before use.

The surface of metal-oxide semiconductors (MOS) thin films was characterized by X-ray photoelectron spectroscopy (XPS; AXIS-NOVA, Kratos, Inc.) in an ultrahigh-vacuum chamber (4.2×10^{-9} Torr). The sample was prepared on a silicon dioxide layer on silicon wafer (Si/SiO_2). The results were obtain by using a monochromatic Al Kα ($h\nu$ = 1486.6 eV) source. The binding energy shift was corrected using the C 1s peak, and before analysis, surface was etched by an Ar+ gun (1 kV, 1 min).

Device Fabrication. TFTs were fabricated on thermally grown 100 nm silicon dioxide layers on silicon wafer (Si/SiO_2), which is used as bottom-gate electrode/dielectric, respectively. Silicon wafers were cleaned in deionized water, acetone, and isopropanol with ultrasonic baths, sequentially. After sonication, residual solvents were removed in an oven heated at 110 °C for 30 min, and then, the samples were treated by UV ozone for 20 min. Thereafter, film deposition was spin-coated: the first step at 500 rpm for 5 s and the second at 2000 rpm for 30 s. After that, the samples were annealed at 100 and 400 °C sequentially for different durations. The details of the fabrication were as follows: first annealing at 100 °C while supplying saturated water vapor in a closed container for 3−5 min; second annealing was done at 400 °C for a time ranging from 5 to 60 min. Finally, source and drain electrodes of 50 nm thick Al were deposited by thermal evaporation via shadow mask. The channel length and width are 100 and 1000 μm, respectively. Electrical characterizations were performed in an N_2-purged glovebox using a Kithely-4200 semiconductor parameter analyzer.

3. RESULTS AND DISCUSSION

Figure 1 shows the formation process of metal-oxide film using the conventional thermal annealing or water vapor annealing (WVA). The metal-oxide precursor essentially necessitates a high-temperature annealing (>350 °C) for decomposition of the bonds in the precursor and the formation of new metal and oxide bonds without the need for additional oxidization agent [Figure 2].[8,11] We thus employed nitrate-based metal salt as

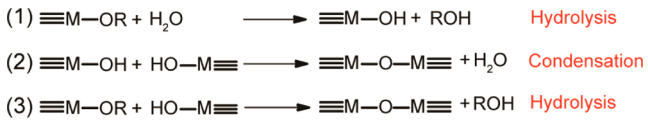

Figure 2. Chemical reaction steps of precursor oxidation.

precursor solution and 2-methoxyethanol (2-ME) as a solvent. Nitrate metal salt can be easily dissolved in 2-ME solvent to form metal alkoxide (M−OR) at room temperature. To obtain metal-oxide film, the alkoxide molecules undergo two major reactions, i.e., hydrolysis and condensation, sequentially.

For the sake of efficient decomposition of metal alkoxide, enough H_2O molecules should be supplied to the MOS precursor film because the metal alkoxides change to oxide with H_2O molecules at the first hydrolysis reaction (Figure 2). Therefore, deficiency of H_2O molecules can result in a stop at the initial stage of the overall reaction and rapidly decrease the total reaction efficiency. In the hydrolysis reaction, metal alkoxides are initially reacted with oxygen atoms in H_2O molecules; therefore, the reaction efficiency strongly depends on the partial pressure of H_2O molecules in air. However, it is

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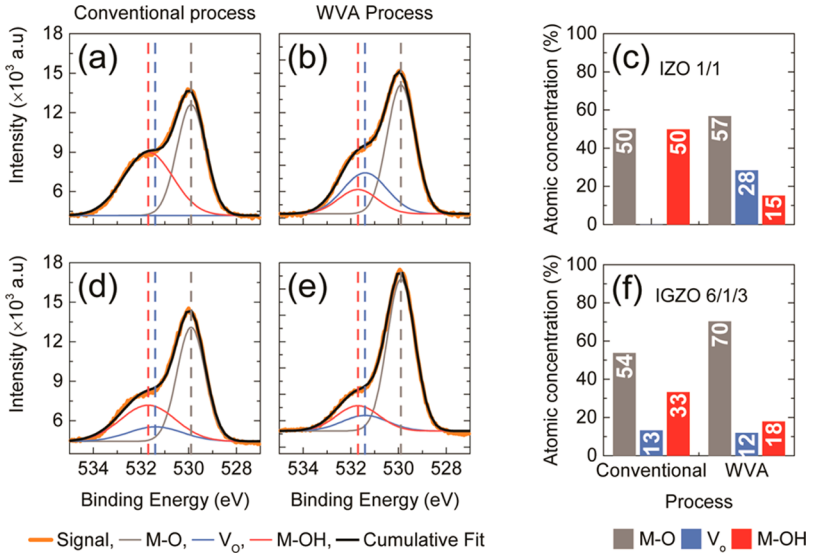

Figure 3. XPS results of O 1s peaks for the IZO (1/1) and IGZO (6/1/3) films. (a) and (d) show the conventionally processed and (b) and (e) are for stage 1 of WVA processed films. (c) and (f) show the O 1s peak-based component concentration for M−O, V_O, and M−OH.

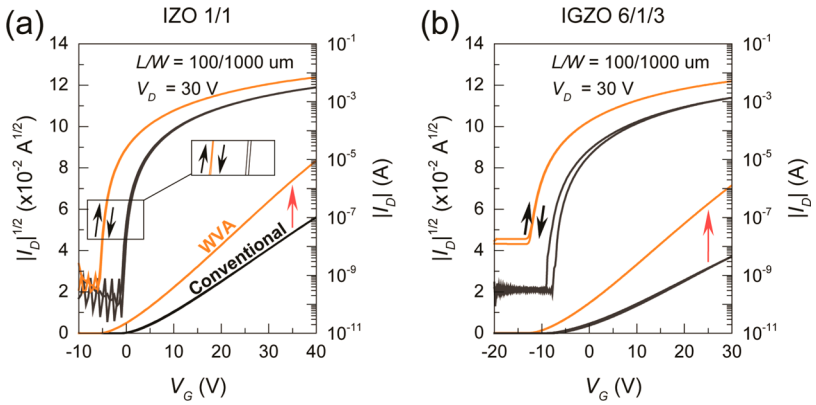

Figure 4. Transfer characteristics of the IZO (1/1) and IGZO (6/1/3) TFTs with different processing methods where all precursors with water vapor annealing show enhanced electrical performance.

very difficult to obtain well-distributed H_2O molecules via conventional thermal annealing since such a high temperature (>100 °C) quickly vaporizes the moisture in the air surrounding the sample, thereby relatively fewer H_2O molecules are provided. Unlike the convention thermal annealing process, the WVA process continually supplies excess H_2O molecules to the AOS precursor film during the whole prebaking process (at 100 °C for 3−5 min) by vaporization of water from a water reservoir (see Figure 1). This excess supply of H_2O molecules sustains the moisture concentration at a constant level and leads to an efficient hydrolysis reaction.

Figure 3 illustrates the X-ray photoelectron spectroscopy (XPS) data and calculated atomic concentrations of the thermally annealed and water-vapor annealed IZO and IGZO thin films. For XPS analysis, MOS films were prepared on p-doped silicon substrates and volume mixed with a 0.1 M concentration precursor of IZO (In/Zn = 1:1) and IGZO (In/Zn/Ga = 6:1:3), respectively. The precursor was mixed from each 0.1 M concentrate mother solution of In, Zn, and Ga nitrate salt dissolved in 2-methoxyethanol by volume ratio. The XPS data show that the O 1s peak is composed of three main components: metal oxide (M−O, 529.9 eV), oxygen vacancy (V_O, 531.4 eV), and metal hydroxide (M−OH, 531.7 eV). The signal of O 1s can be thus divided into three major components with different Gaussian distributions and then be integrated to fit the experimental result so as to get each atomic concentration. To investigate the effects on decomposition processes of adding water vapor of the nitrate precursor, we separate the entire annealing into two stages: stage 1 is a mild annealing process (100 °C for 1−5 min) for the hydrolysis reaction and stage 2 is a hard annealing process (400 °C for 5−60 min) for densification of the film.[12] Generally, the performance and stability of metal oxide TFTs strongly depends on three atom components which are metal-oxide (M−O), metal hydroxide (M−OH), and oxygen vacancy (V_O).[7,13] As shown in Figure 3b,e, water vapor annealed IZO (1/1) and IGZO (6/1/3) films exhibit larger M−O and smaller M−OH atomic concentrations than the conventionally processed films. In particular, metal hydroxide (M−OH) atom concentration dramatically decreases from 50% to 15% in IZO (1/1) film and from 33% to 18% in IGZO (6/1/3) film, whereas M−O concentration increases from 50% to 57% for IZO (1/1) film and from 57% to 70% for IGZO (6/1/3) film, respectively. This result indicates that the WVA process effectively promotes the transformation from metal alkoxide

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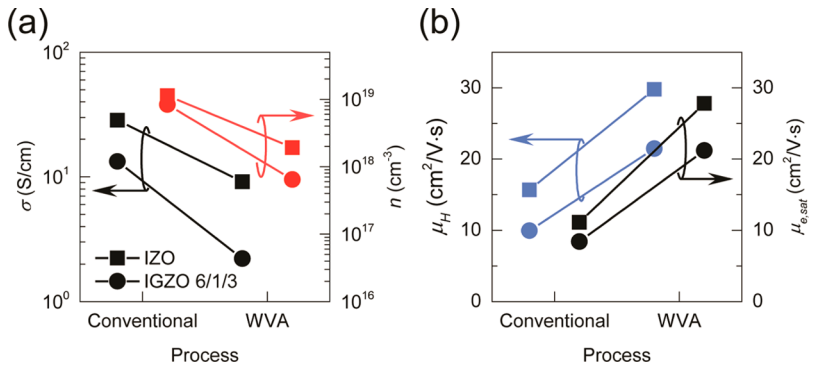

Figure 5. Results of Hall Effect measurements: (a) film conductivity (σ), carrier concentration (n), (b) Hall mobility (μ_H), and effective saturation mobility ($\mu_{e,sat}$) of the conventional and WVA processed MOS films.

and hydroxide into metal oxide during the hydrolysis process (stage 1).

The performance of water-vapor annealed IZO and IGZO TFTs are consistent with the XPS results. Figure 4 and Figure S2 in the Supporting Information show transfer characteristics of the IZO and IGZO TFTs fabricated by using WVA and conventional annealing processes. Those metal-oxide TFTs were fabricated with a bottom-gate and top-contact device structure and based on semiconducting IZO (1/1) and IGZO (6/1/3) films which display n-type device properties. The conventionally processed IZO (1/1) and IGZO (6/1/3) TFTs exhibit a saturation mobility of 11.1 and 8.4 $cm^2/V{\cdot}s$, respectively, whereas the WVA processed IZO (1/1) and IGZO (6/1/3) counterparts show significantly improved mobility up to 27.8 and 21.2 $cm^2/V{\cdot}s$, respectively. This means that the electron mobility in WVA processed TFTs is improved more than a factor of 2, correlated to the XPS result of atom concentration that is discussed above. Therefore, supplying additional water molecules through WVA can effectively assist the alkoxide precursors to convert into semiconducting metal oxides. In particular, WVA processing effectively works at the low-temperature annealing stage (hydrolysis process) for efficient decomposition reaction since the nitrate precursor starts to decompose at relatively low temperature (100−200 °C).[14]

To better understand the underlying mechanism, we performed Hall-effect measurements on the films with and without WVA. Figure 5a shows that the film conductivity and the free electron concentration are all reduced after WVA. This is consistent with the previous result in which the presence of H_2O molecules during WVA promotes oxide formation, which decreases the doping effects arising from donor states (the next stability test confirms this inference). Figure 5b illustrates the Hall mobility (μ_H) in which the effective mobility extracted from the current−voltage characteristics ($\mu_{e,sat}$) is also incorporated for comparison. One can find that μ_H exhibits a similar rising tendency to $\mu_{e,sat}$ after WVA. Note that μ_H represents the intrinsic electron mobility in the bulk of the oxide semiconductor because only the moving electrons experience the Lorentz force and thereby accumulate to form the Hall voltage, whereas $\mu_{e,sat}$ is an apparent (or averaged) mobility of the overall electrons that transport at the interface between the oxide semiconductor and the gate dielectric. The increased mobility after WVA, combined with the reduced conductivity and carrier concentration, signify that the charge transport in the conventionally processed MOS films is limited by scattering, perhaps owing to defect centers and the electrons localized in donor states.

Moreover, the TFTs based on these highly oxidized MOS films demonstrate better bias stability relative to those with the conventionally processed MOS. Figures 6 and S5, Supporting

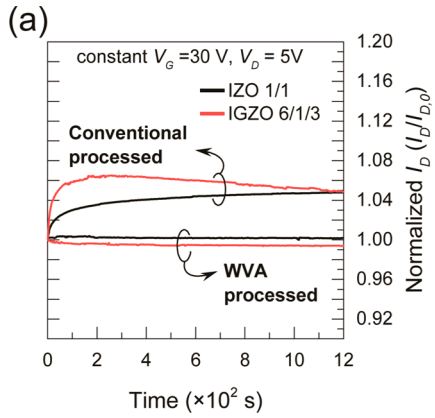

Figure 6. (a) Bias stress test of FETs-based IZO 1/1 and IGZO 6/1/3 precursor MOS films. The normalized current decay for devices performed on time under the constant biases of V_G = 30 V and V_D = 5 V.

Information, show a comparison of two such TFTs by means of the normalized on-state current (I_D) versus the biasing time (at V_G = 30 V, V_D = 5 V). A significant increase of I_D (>5%) is quickly observed for the conventionally processed MOS (IZO and IGZO), yet great stability with a tiny change of I_D (<1%) over 1000 s is obtained for the WVA-processed counterparts. The poor stability of the conventionally processed MOS would be attributed to the incomplete oxidation or more metal hydroxide, i.e., donor states.[15] More donor states can be created upon bias stressing, thus increasing the on-state current. The greatly improved stability implies that such defects are effectively eliminated by WVA

Figure 7 and Table 1 summarize the extracted device parameters. It can be readily seen that the performance of the WVA-processed TFTs is considerably enhanced as compared with the conventional ones. Even with the same processing time, mobilities of IZO (1/1) and IGZO (6/1/3) TFTs are increased from 11.1 and 8.4 $cm^2/V{\cdot}s$ to 27.8 and 21.2 $cm^2/V{\cdot}s$, respectively. Even though WVA process requires high annealing temperature (stage 2), if processing time was reduced from 60 to 5 min, WVA-processed devices still show two times higher mobility than the conventionally long-time processed ones. For

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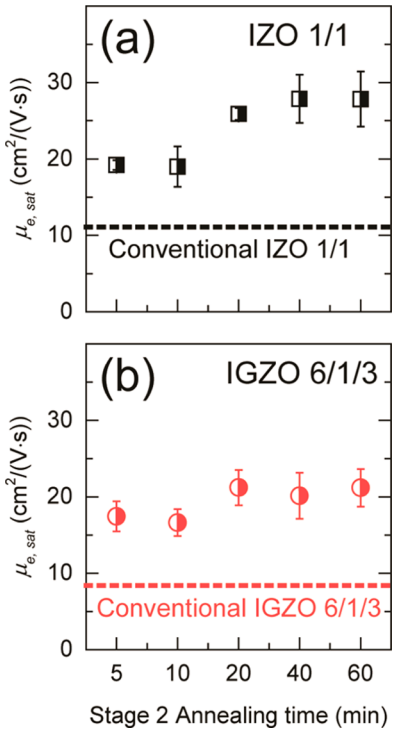

Figure 7. Trend of mobilities of (a) IZO (1/1) and (b) IGZO (6/1/3) according to annealing time.

instance, the IZO (1/1) TFTs's mobility increases from 11.1 to 19.2 cm^2/V·s, and the IGZO (6/1/3) TFTs's mobility rises from 8.4 to 17.4 cm^2/V·s as shown in Figure 7. Indeed, well-activated initiation of the precursor reaction and rapid reaction completion by using WVA can considerably reduce the annealing time to only a few minutes. Furthermore, we attempted this WVA method with many other precursors and always obtained greatly improved performance (see Figure S4, Supporting Information). This result indicates that WVA is an effective and versatile method for rapidly producing high-performance metal-oxide devices based on sol−gel.

4. CONCLUSIONS

In conclusion, we demonstrate water vapor annealing (WVA) as a simple and versatile method for producing high-performance semiconducting metal oxide by using facile solution-based processes. It can be done in atmospheric conditions within a few minutes and so is very well suited to rapid manufacturing for practical applications. We found that the performance of WVA-processed TFTs is considerably enhanced with respect to conventionally processed counterparts, and the performance enhancement remains significant even if the WVA time is reduced to a few minutes. In examination of XPS results, it turns out that the constantly supplied water activates the initial reaction of precursor decomposition and greatly improves the reaction efficiency; therefore, the entire process can be completed in a short time. Moreover, WVA promotes atom organization and densification of the metal oxide film, which is beneficial for charge transport. After several tries using various metal oxides, this method is indeed sufficiently versatile to be applied to oxide TFTs fabrication for extensive applications.

■ ASSOCIATED CONTENT

Supporting Information

XPS results and transfer and output characteristics of IZO (1/1), IGZO (6/1/3), and IGZO (6/1.5/2.5) with conventional and WVA processed MOS FETs. The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsami.5b04374.

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Table 1. Basic Parameters of All of Thermal or Water-Vapor Annealed IZO and IGZO TFTs

process	stage 1 time[a] (min)	stage 2 time[a] (min)	precursor	$\mu_{e,sat}$ (cm^2/V·s)	$V_{e,TH}$ (V)	S.S (V/dec.)	I_{on}/I_{off}
conventional annealing	5 min	60 min	IZO (1/1)	11.10	3.0	0.6	1.2×10^7
			IGZO (6/1/3)	8.4	−2.4	0.5	1.2×10^7
			IGZO (6/1.5/2.5)	4.8	2.9	0.6	1.6×10^7
	5 min	60 min	IZO (1/1)	27.8	−5.5	1.0	2.1×10^5
			IGZO (6/1/3)	21.2	−5.9	1.3	2.1×10^5
			IGZO (6/1.5/2.5)	15.1	−4.5	0.9	2.1×10^6
		40 min	IZO (1/1)	27.9	−7.7	1.9	4.7×10^5
			IGZO (6/1/3)	20.1	−8.1	1.9	4.7×10^5
			IGZO (6/1.5/2.5)	18.3	−8.4	1.7	2.2×10^5
water vapor annealing (only stage 1)		20 min	IZO (1/1)	25.9	−6.5	2.0	2.0×10^5
			IGZO (6/1/3)	21.2	−10.2	1.8	2.0×10^5
			IGZO (6/1.5/2.5)	16.3	−8.7	1.8	2.7×10^5
	3 min	10 min	IZO (1/1)	19.0	−4.6	1.7	1.7×10^5
			IGZO (6/1/3)	16.6	−0.7	1.8	5.3×10^4
			IGZO (6/1.5/2.5)	15.7	−2.4	2.1	1.1×10^5
		5 min	IZO (1/1)	19.2	−1.6	2.8	1.5×10^4
			IGZO (6/1/3)	17.4	−1.8	1.7	1.0×10^5
			IGZO (6/1.5/2.5)	16.4	−1.4	2.1	2.7×10^4

[a]Annealing processes for stage 1 and stage 2 were performed at 100 and 400 °C, respectively.

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Notes

The authors declare no competing financial interest.

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