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# Bias and illumination instability analysis of solution-processed a-InGaZnO thin-film transistors with different component ratios



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# ABSTRACT

We investigated the effect of In and Zn component ratios on an amorphous indium-gallium-zinc-oxide (a-IGZO) layer in oxide thin-film transistors (TFTs). Different types of stress including negative bias, positive bias, negative bias illumination stress, and instability of a-IGZO TFTs were compared. Owing to the difference in the composition ratio of In:Ga:Zn (5:1:4, 6:1:3, 7:1:2), the respective devices had threshold voltages of 7, 5, and 4.5 V. The high content of In in a-IGZO leads to a higher concentration of oxygen vacancy, which results in sensitivity toward illumination stress. The above results indicate that the threshold voltage shift is primarily due to the trapping of electrons in the traps located in the interfaces or dielectric layers. We analyzed the IGZO thin film properties using UV–visible, X-ray diffraction, and X-ray photoelectron spectroscopy measurements.

# 1. Introduction

Amorphous indium-gallium-zinc-oxide (a-IGZO) thin-film transistors (TFTs) have attracted considerable attention as promising alternatives to amorphous silicon (a-Si) and poly-crystalline silicon (poly-Si) for use as the backplane in active-matrix displays. The a-IGZO TFTs have better electrical characteristics—high field-effect mobility ( $\mu_{FET}$ ), low sub-threshold swing (S.S.), low threshold voltage ( $V_{th}$ ), and high on/off ratio—compared to a-Si TFTs, owing to the higher symmetry of s-orbitals in heavy metal ions and higher transparency derived from a relatively large band gap [1–3]. The a-IGZO film based on sol-gel chemistry is of particular interest, because a solid metal oxide material with excellent electrical characteristics can be easily synthesized from small molecular precursors using simple solution processes [4].

The indium component of the a-IGZO sol-gel solution is necessary to improve the electrical performance of the TFTs, because heavy metal indium cations share electrons in 5s orbitals and act as electron pathways, which contributes to an increase in the carrier mobility of TFTs [5,6]. Zinc-oxide-based transparent semiconductors are applicable to the sol-gel process, which makes low-cost and printable manufacturing possible [7]. The role of individual metal elements in multi-component oxide semiconductor systems [8,9] in the formation of thin films with high performance and at low temperature for flexible devices [10,11],

and stability against bias, temperature, and the presence of ambient gasses [12–13] affect the functionality of a-IGZO TFTs.

Various stresses on a-IGZO TFTs are responsible for the threshold voltage shift ( $\Delta V_{th}$ ). The instability of  $V_{th}$  of oxide TFTs under gate voltage stress originates from the gate-field-induced molecular adsorption/desorption of oxygen, hydrogen, or water on the back channel surface and the interface between the channel and gate insulator (G.I.). The charge trapping phenomena of the G.I. results in  $V_{th}$  instability [14]. The sol-gel methods used in the solution process cause structural defects such as pores or pin holes through volatilization during the annealing process. Moreover, organic residues can easily be captured by these structural defects and act as impurities after they are trapped in the thin-film bulk. Solution-processed a-IGZO TFTs have two typical defects: intrinsic oxygen vacancy defects (Vo) and organic-chemicalinduced defects [15,16]. These exposed oxygen vacancy defects are either double  $(Vo^{2+})$  or single  $(Vo^{+})$  ionized by photo-generated holes in shallow donors. The organic chemicals induce defects, such as pores, pinholes, and organic residues [17]. This complexity of the oxide TFT system has prevented unambiguous and in-depth studies from being conducted to determine the exact relationship between the channel material properties (channel composition and film density) and the resulting transistor instability.

Therefore, in this study, we fabricated a-IGZO TFTs using sol-gel

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Fig. 1. (a) Cross-sectional schematic of the fabricated bottom gate-top contact a-IGZO TFTs. (b) Optical transmittance of the a-IGZO films with three different component ratios. (Inset: Optical energy band gap of the a-IGZO films) (c) GIXRD result of the a-IGZO films formed with various component ratios.

methods. The chemical reactions in the a-IGZO solution derived using sol-gel methods with varying indium and zinc component ratios control the structural and electrical properties of films, and by extension, the device performance. We analyzed the negative bias stress (NBS), positive bias stress (PBS), negative bias illumination stress (NBIS), and positive bias illumination stress (PBIS) in the a-IGZO TFTs. The degradation behavior and mechanism of the device under these various operating conditions were studied.

## 2. Experiment details

We fabricated the bottom gate-top contact source/drain structured a-IGZO TFTs as shown in Fig. 1(a). The a-IGZO solution was prepared by dissolving indium nitrate hydrate, gallium nitrate hydrate, and zinc acetate dihydrate powders (Sigma Aldrich, USA) in 2-methoxyethanol at a molar concentration of 0.1 M. We prepared three different solutions with component ratios of 5:1:4, 6:1:3, and 7:1:2 (In:Ga:Zn). The mixtures were stirred at the temperature of 75 °C for 12 h to facilitate the dissolving process and to stabilize the solution. Heavily doped silicon wafer was used both as a substrate and as a gate electrode for the fabrication of TFT. A silicon dioxide layer was deposited on the Si using plasma enhanced chemical vapor deposition process at a maximum temperature of < 200 °C. The substrates were cleaned in an ultrasonic bath with acetone, isopropyl alcohol, and deionized water for 20 min each. The UV-ozone treatment was applied on the surface of SiO<sub>2</sub> for 30 min before spin coating to remove the unnecessary organics. The a-IGZO was filtered through a syringe filter and spin-coated at a speed of 4000 rpm on the SiO<sub>2</sub> substrate for 30 s. The deposited films were subsequently annealed at 200 °C for 10 min on a hot-plate under air ambient. The annealed a-IGZO films were patterned using photolithography via a wet etch process. The wet etching of the IGZO layer was carried out using LCE-12K (LCE etchant was mixed with 3 vol% of deionized water) from Cyantek Corporation. After patterning the a-IGZO layer, the films were annealed at the temperature of 350 °C for 3 h on a hot plate. The thickness of the a-IGZO film in this case was 10 nm  $(\pm 2 \text{ nm})$ . The Al source-drain electrodes (100 nm), were deposited using a shadow mask via thermal evaporation. The oxide TFTs thus obtained had a channel length (L) and width (W) of 100 and 1000 µm, respectively. The electrical characteristics were investigated using an Agilent 4145B semiconductor parameter analyzer. The surface morphology and roughness of the solution-processed a-IGZO films were examined using atomic force microscopy (AFM) (XE-100, Park system) in the non-contact mode. The crystallographic orientation of the films were examined using X-ray diffraction (XRD) (D8 advance, Bruker Corporation) with the acceleration voltage of 40 kV and emission current of 150 mA using a Cu-Ka source. All measurements were performed in reflection geometry (Bragg-Brentano) as coupled  $\theta$ -2 $\theta$  scans with a step size of 0.02°. The surface chemical composition of the films was measured using X-ray photoelectron spectroscopy (XPS) (Escalab 250, Thermo) with an Al-K $\alpha$  source and beam spot diameter of 300  $\mu$ m. The peaks were fitted and analyzed using Thermo Avantage software in details using Gaussian/Lorenzian (with Lorentzian/Gaussian ratio of O1s 30%) mixture. Surface composition and concentrations were calculated from appropriate peak area. Binding energy scale was corrected based on C<sub>1s</sub> peak from contaminations (around 284.8 eV) as the internal binding energy standard. The optical properties of a-IGZO films were examined using a UV/vis spectrophotometer (Shimadzu UV-1601). The entire fabrication and measurement processes were performed in ambient air.

# 3. Results and discussion

Fig. 1(b) shows the transmittance of a-IGZO films, which typically exhibit a transmittance of > 90% in the visible range (300–800 nm). The inset of Fig. 1(b) shows the optical band gap of the a-IGZO film [18,19]. In order to identify the optical absorption coefficient ( $\alpha$ ) of the a-IGZO films, we used Lambert's law:

$$\alpha = \frac{1}{d} \ln \left( \frac{1}{T} \right),$$

where *d* is the film thickness and *T* is the transmittance. In a direct-transition semiconductor material, the absorption coefficient  $\alpha$  is correlated to the optical band gap according to the following equation:

$$(\alpha \hbar \upsilon)^2 = A(\hbar \upsilon - E_g),$$

where  $\hbar$  is Planck's constant,  $\nu$  is the frequency of the incident photon, A is a constant that depends on the mobility of the electrons and holes in the material, and  $E_g$  is the optical band gap. Hence, the optical band gap of the a-IGZO films was determined using Tuac's plot, i.e., the linear part of the  $(\alpha\hbar\nu)^2$  curve was extrapolated toward the energy  $\hbar\nu$  axis at

Fig. 2. AFM image of the a-IGZO films formed with various component ratios (In:Ga:Zn) (a) 5:1:4 (b) 6:1:3 (c) 7:1:2.



 $(\alpha\hbar\nu)^2 = 0$ . The obtained band gap of the a-IGZO films is approximately 3.5 eV, as shown in Fig. 1(b). The band gap for all the three component ratios was calculated and determined to be the same (3.5 eV).

The amorphous nature of the a-IGZO film can be confirmed using grazing incidence X-ray diffraction (GIXRD). As shown in Fig. 1(c), the same In peak (2, 2, 2) of all the three layers indicates that the crystallizations in the a-IGZO film are the same regardless of the composition ratio [20]. In the GIXRD measurement of the IGZO film, Chang et al. proposed that a weak broad peak at 33–34° can be attributed to an amorphous-like nanocrystal phase [21]. The amorphous-like phase was not a fully amorphous phase but an amorphous phase with a local small crystalline phase. As shown in Fig. 2, the surface morphology of a-IGZO films with different In and Zn ratios of 5:1:4, 6:1:3, and 7:1:2 exhibits a root-mean-square roughness of 0.05, 0.051, and 0.034 nm, respectively. The three devices exhibited similar coarse morphology and roughness. The AFM image data support the GIXRD and transmittance data corresponding to Fig. 1(c).

The measured I–V characteristics of the a-IGZO TFTs with various In and Zn components using an Agilent 4145B semiconductor parameter analyzer are shown in Fig. 3. The transistors were analyzed to extract their electrical properties. The mobility was extracted in the saturation regime of the TFTs using the following equation:



Fig. 3. Effect of In and Zn component ratio on the electrical characteristics of the a-IGZO TFTs.

$$\mu_{sat} = \left(\frac{\partial \sqrt{I_{DS}}}{\partial V_{GS}}\right)^2 \times 2\frac{L}{CW}$$

where  $\mu_{sat}$  is the saturation mobility,  $I_{DS}$  is the drain current,  $V_{GS}$  is the

#### Table 1

I-V characteristics of a-IGZO TFTs with three different component ratios.

In:Ga:Zn	Mobility [cm <sup>2</sup> /V·s]	V <sub>th</sub> [V]	I <sub>on/off</sub> ratio	S [V/decade]
5:1:4	1.18	7	$> 10^8$	1.36
6:1:3	1.44	5	> 10^8	1.38
7:1:2	1.62	4.5	> 10^8	1.42

gate voltage, L is the channel length, W is the channel width, and C is the gate oxide capacitance.

The transfer characteristics of the a-IGZO TFTs was measured when  $V_{GS}$  was swept from -20 to 40 V with a sweep step of 0.5 V at  $V_{DS}$  of 20 V. The device with lower In component (5:1:4) exhibited  $V_{th}$  of 7 V and  $\mu_{FET}$  of 1.18 cm<sup>2</sup>/V·s. The device with higher In component (7:1:2) exhibited  $V_{th}$  of 4.5 V and  $\mu_{FET}$  of 1.62 cm<sup>2</sup>/V·s. The value of  $\mu_{FET}$  increased monotonically with the increase in the In<sub>2</sub>O<sub>3</sub> content in the IGZO channel layer, and  $V_{th}$  shifted to the negative voltage direction. The physical role of the In content has been identified as the mobility enhancer and carrier generator owing to its 5s orbital radius [7,8]. Moreover, at higher In concentrations, the In<sup>3+</sup> ions change to InO<sub>6</sub> octahedron structure. The performances of all the a-IGZO TFTs are summarized in Table 1.

Fig. 4 shows the XPS measurements of the a-IGZO films with different component ratios. The black curves are X-ray photoelectron spectra (O (1s) peak) of thermally annealed a-IGZO films. The peak fit analysis of the spectra shows the contributions of peaks at approximately 530.0 (red), 531.5 (blue), and 532.5 eV (pink), which are due to oxygen atoms in the M-O lattice, oxygen atoms near oxygen vacancies and in M-OC bonds, and oxygen atoms in M-OH compounds, respectively. The XPS analysis of all the a-IGZO films is summarized in Table 2. In the deposition of the multicomponent metal oxide thin films, the preferential bonding characteristics between the metal positive ion and negative oxygen ion can usually be explained by the electronegativity difference, which is a measure of the ability of an element to attract electrons [22]. The electronegativity differences (usually called  $\Delta$ EN) for In–O, Ga–O, and Zn–O bonds are 1.78, 1.81, and 1.65, respectively. From the table, we can observe that M-O bonding does not change significantly for all the component ratios. However, the increased In proportion and the reduction in the Zn proportion lead to a higher percentage of oxygen vacancies and less M-OH bonding. Thus, we can assume that In metals attract an oxygen ion more strongly as compared to a Zn metal and thus, it is expected that Zn–O bonding is a relatively reduced metal-oxygen bond for the low concentration of IGZO (4:1:3) [22].

We performed the PBS or NBS under a gate bias  $V_G$  of 20 or -20 V with different time duration, and we measured the device performance under a drain bias  $V_D$  of 20 V. The rate of change of  $V_{th}$  was calculated from 0 s to 3000 s. The performances of all the a-IGZO TFTs under the PBS and NBS are summarized in Table 3.

Fig. 5(a) shows the threshold voltage shift of the devices with various In and Zn component ratios in the active layer in relation to the stress time under the PBS and NBS conditions. Fig. 5(a) indicates that the shift in  $V_{th}$  is attributed to two types of charge trap: hole trap and electron trap. The PBS showed that the device exhibited a smaller  $V_{th}$ variation and higher stability when the In proportion increased and Zn proportion decreased. However, NBS showed that, when the In proportion decreased and Zn proportion increased, the devices showed smaller V<sub>th</sub> variation and higher stability. The threshold voltage shift owing to PBS follows the same mechanism as that of a general n-type a-IGZO TFT. Under positive gate bias, the trapped electrons at the interface between the active layer and gate insulator reduce the effective applied gate voltage, leading to a positive shift in  $V_{th}$  [23]. In particular, for a component ratio of 5:1:4,  $V_{th}$  shifts more drastically to the positive side owing to the increased M-OH bonding, following the mechanism shown in Fig. 5(c). However, for NBS, as n-type a-IGZO TFTs have



Fig. 4. X-ray photoelectron spectra (O(1s) peak) of thermally annealed a-IGZO films with the component ratios of (a) 5:1:4, (b) 6:1:3, and (c) 7:1:2.

Table 2 XPS (O1s peak) measurement of a-IGZO films with three different component ratios.

In:Ga:Zn	M-O [%]	V <sub>O</sub> [%]	M-OH [%]
5:1:4	52.49	33.91	13.6
6:1:3	53.97	34.53	11.5
7:1:2	54 63	35.87	9.5

negligible holes in the valence band, hole trapping in the gate insulator or at the interface between the semiconductor and gate dielectric is extremely difficult, even under negative gate bias [24]. The component ratio of 7:1:2 showed a more significant negative shift owing to a relatively larger number of oxygen vacancy bonds (Fig. 5(c)). From the results, it can be observed that the device with a ratio of 7:1:2 is more stable under PBS owing to the decreased effect of M—OH, and the device with a ratio of 5:1:4 is more stable under NBS owing to the decreased effect of the oxygen vacancies.

We performed the PBIS or NBIS under a gate bias  $V_G$  of 20 or -20 V with different time duration and measured the performance under a

#### Table 3

In:Ga:Zn	0 s	10 s	30 s	100 s	300 s	1000 s	3000 s
Positive bias st	ress $\Delta V_{th}$ [V]						
5:1:4	0 [V]	+ 0.5 [V]	+ 1.6 [V]	+ 2.4 [V]	+ 3.6 [V]	+ 5.3 [V]	+ 7.9 [V]
6:1:3	0 [V]	+ 0.2 [V]	+ 1[V]	+ 2.1 [V]	+ 3.1 [V]	+ 4.4 [V]	+ 7.5 [V]
7:1:2	0 [V]	+0.1 [V]	+ 0.5 [V]	+ 1.3 [V]	+ 2.5 [V]	+ 3.2 [V]	+ 5.5 [V]
Negative bias s	tress $\Delta V_{th}$ [V]						
5:1:4	0 [V]	0 [V]	0 [V]	0 [V]	-0.1 [V]	-0.3 [V]	-0.4 [V]
6:1:3	0 [V]	0 [V]	0 [V]	-0.1 [V]	-0.2 [V]	-0.4 [V]	-1.4 [V]
7:1:2	0 [V]	0[V]	0 [V]	-0.1 [V]	- 0.3 [V]	-0.9 [V]	- 2.6 [V]

Effect of In and Zn component ratios (In:Ga:Zn = 5:1:4, 6:1:3 and 7:1:2) on the threshold voltage shift of the a-IGZO TFTs with positive bias stress and negative bias stress.

drain bias  $V_D$  of 20 V. During the application of PBIS and NBIS, light illumination and gate voltage were applied together for 1000 s. The performances of all the a-IGZO TFTs under PBIS and NBIS are summarized in Table 4.

Fig. 5(b) shows the threshold voltage shift of the devices with various In and Zn component ratios in the active layer in relation to the light-intensity-dependent PBIS and NBIS. From the results, for the component ratios of 5:1:4 and 6:1:3, when the illumination stress was < 10 W/m<sup>2</sup>, the effect of PBS was dominant. When the illumination stress exceeded 10 W/m<sup>2</sup>, the illumination stress effect became dominant for a change in  $V_{th}$ . However, for the component ratio of 7:1:2, when the illumination stress exceeded 5 W/m<sup>2</sup>,  $V_{th}$  changed drastically, which indicated that the illumination stress was dominant. The devices were more stable under PBIS when the Zn proportion

increased and In proportion decreased. However, NBIS showed that, for the component ratios of 5:1:4 and 6:1:3,  $V_{th}$  showed a smaller change compared with that for the component ratio of 7:1:2, when the illumination stress was smaller than 10 W/m<sup>2</sup>.  $V_{th}$  reached a saturation value of -18 V under 25 W/m<sup>2</sup> for all the component ratios. The mechanism of NBIS is explained in Fig. 5(c). Under PBIS, even though the holes are not captured as in the NBS condition, the illumination stress still leads to a small shift in  $V_{th}$  because the shallow states that exist in the a-IGZO film generate an electron-hole pair. However, during NBIS, the photo-generation of electron-hole pairs occurs and positive charges drift toward the gate insulator/active layer interface. The existing neutral oxygen vacancies (V<sub>O</sub>) at the deep levels are ionized by hole capture (forming either V<sub>O</sub><sup>+</sup> or V<sub>O</sub><sup>2+</sup>). The ionization of V<sub>O</sub> results in the formation of defects (V<sub>O</sub><sup>+</sup> and/or V<sub>O</sub><sup>2+</sup>) and the trapping of



Fig. 5. Effect of In and Zn component ratios (In:Ga:Zn = 5:1:4, 6:1:3, and 7:1:2) on the threshold voltage shift of the a-IGZO TFTs with (a) bias stress (b) illumination stress. (c) The schematics of the energy-level diagrams with various stresses (PBS, NBS, PBIS, and NBIS).

#### Table 4

Effect of In and Zn component ratios (In:Ga:Zn = 5:1:4, 6:1:3 and 7:1:2) on the threshold voltage shift of the a-IGZO TFTs with positive bias illumination stress or negative bias illumination stress at the gate voltage of 20 or -20 [V] for 1000 s.

In:Ga:Zn	0 [W/ m <sup>2</sup> ]	5 [W/m <sup>2</sup> ]	1 [W/m <sup>2</sup> ]	15 [W/m <sup>2</sup> ]	20 [W/m <sup>2</sup> ]	25 [W/m <sup>2</sup> ]	
Positive bias illumination stress $\Delta V_{th}$ [V]							
5:1:4	0 [V]	+ 2.3 [V]	+ 2.5 [V]	+ 2.2 [V]	+1.4 [V]	-1.2 [V]	
6:1:3	0 [V]	+ 2.2 [V]	+ 2.3 [V]	+1.9 [V]	-0.5 [V]	– 2.5 [V]	
7:1:2	0 [V]	+ 1.6 [V]	+1.3 [V]	-0.4 [V]	– 2.5 [V]	– 4.8 [V]	
Negative bias illumination stress $\Delta V_{th}$ [V]							
5:1:4	0 [V]	- 1.2 [V]	– 5.2 [V]	– 14.5 [V]	– 19.6 [V]	– 21.8 [V]	
6:1:3	0 [V]	– 1.8 [V]	- 8.1 [V]	-18.1 [V]	– 21.7 [V]	– 22.7 [V]	
7:1:2	0 [V]	– 3.6 [V]	– 12.6 [V]	- 19.4 [V]	– 22.5 [V]	– 23.3 [V]	

these ionized oxygen vacancies and positive charges at the active-layer/gate-insulator interface explains the negative  $V_{th}$  [25].

The electron and hole trapping mechanisms act in opposition to each other. Furthermore, the efficacy of these two trapping mechanisms strongly depends on the oxygen content in the a-IGZO bulk. The occupied electron trapping states (acceptor-like) were formed more easily when the oxygen vacancy content was high, and the unoccupied hole-trapping states (donor-like) were formed more easily when the M–OH content was high. Fig. 5(c) shows the energy band diagram of the a-IGZO TFT under various stress conditions. The accumulated electrons near the dielectric interface were trapped by shallow acceptor-like trap states, which were oxygen-related defect states. The depleted holes near the dielectric interface were trapped by deep donor-like trap states, which were M–OH defect states. During the application of various stresses, oxygen ions ( $O^{2-}$ ; electron trapping state) or oxygen vacancies ( $V_O^{2^+}$ ; hole trapping state) may have been collected at the dielectric surface.

Based on these results, we present stable and reliable a-IGZO TFTs, when compared with other solution-processed devices. The generation of interface traps, trapped oxygen vacancy, and M—OH concentration during the application of various stresses alters the threshold voltage shift and degrades the reliability of a-IGZO TFTs. The In:Ga:Zn with a ratio of 7:1:2 showed more stability under the PBS owing to a lower OH concentration, whereas the device with a component ratio of 5:1:4 was more stable under illumination stress owing to the lower oxygen vacancy. The proposed a-IGZO TFTs can be used in electrical device circuits for higher performance and better reliability.

#### 4. Conclusion

The analysis of various In and Zn components in a-IGZO for use as an active layer in TFTs was investigated. The device with lower In component (5:1:4) showed  $V_{th}$  of 7 V and  $\mu_{FET}$  of 1.18 cm<sup>2</sup>/V·s. The device with higher In component (7:1:2) showed  $V_{th}$  of 4.5 V and  $\mu_{FET}$  of 1.62 cm<sup>2</sup>/V·s. The effect of PBS, NBS, PBIS, and NBIS on the a-IGZO TFTs was studied in addition. The high percentage of In in a-IGZO leads to a high concentration of oxygen vacancy, which results in the sensitivity toward illumination stress. Consequently, the devices with the component ratios of 6:1:3 and 5:1:4 exhibited more stable characteristics than those with the component ratio of 7:1:2.

## Author contributions

J.-H. Kim and E.-K. Park contributed equally to this work.

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