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Effect of In-Ga-Zn-O active layer channel composition on process temperature for flexible oxide thin-film transistors

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In-Ga-Zn-O (IGZO)-channel oxide thin-film transistors (TFTs) were fabricated on flexible polyethylene naphthalate (PEN) substrates. A lamination and delamination procedure was established that allowed easy handling of the PEN substrate during fabrication. In order to fabricate high-performance flexible IGZO TFTs at lower than normal process temperatures, a 2:1:2 (In:Ga:Zn) IGZO channel composition was proposed. The field-effect mobility, threshold voltage, and subthreshold swing of the fabricated IGZO TFTs were found to be approximately 7.83 $cm^2\ V^{-1}\ s^{-1}$, 1.93 V, and 0.24 V/decade, respectively, even when a final heat treatment was conducted at a temperature as low as 150 °C. The stability characteristics of the devices were also examined under gate bias stress and constant current stress conditions. © *2012 American Vacuum Society*. [http://dx.doi.org/10.1116/1.4731257]

I. INTRODUCTION

Flexible electronic systems implanted on bendable or rollable plastic substrates have great potential in the field of next-generation "consumer electronics." Specifically, flexible-type display panels are the most promising applications of flexible electronic systems. Many groups in both industry and academia have conducted research in the areas of liquid crystal displays and organic light emitting diodes.[1,2] Organic-based semiconducting channels have mainly been proposed as a back-plane device for flexible displays due to their mechanical flexibility.[3] Organic thin-film transistors (TFTs) may be quite suitable for low-cost disposable applications with relatively looser specifications. However, the drawbacks of low field-effect mobility, unsatisfactory ambient stability, and difficulties in integrating the devices with peripheral circuits seriously limit the practical application of organic TFTs. A powerful alternative for improving the device performance of flexible display back-planes is to employ an oxide semiconducting channel in the TFT. The beneficial features of oxide TFTs, such as high carrier mobility, excellent uniformity, and robust device stability, make these structures highly feasible for use as a high-performance back-plane device in flexible displays.[4] Moreover, because the oxide channels are patterned into only small active areas on the substrate, the brittle nature of the oxide film will no longer be a significant problem.

When utilizing oxide TFT back-planes for flexible applications, however, it is critical to ensure both excellent device characteristics and a low process temperature.[5,6] Many approaches have been taken to try to improve both electrical performance and device stability, namely controlling the oxygen partial pressure during deposition,[7–10] the channel composition and material, and the annealing process and ambient conditions[11–13] for the oxide channel layer. The composition of the channel material has an especially large impact on device parameters such as field-effect mobility (μ_{FE}), threshold-voltage (V_{TH}), subthreshold-swing (S.S), and on/off current ratio ($I_{on/off}$ ratio), and also the temperature required to optimize device performance.[14,15] As such, many oxide channel materials and compositions, including In-Zn-O,[13,16,] In-Ga-Zn-O (IGZO),[17,18] Zn-In-Sn-O,[19,20] Al-Zn-Sn-O,[12,21] and Al-In-Zn-Sn-O[22,23] have been proposed and TFTs have been fabricated on glass substrates. All of these materials were confirmed to be effective solutions to obtain good electrical performance. However, the device stability under a bias and/or light illumination could not be guaranteed. We believe that the employment of a suitable composition for the channel layer is one of the most important parameters to solve the requirements of device performance and reliability. Although there have been some encouraging study results on oxide TFTs implemented in flexible displays,[24–26] it is still necessary to satisfy the requirements of device performance and reliability below a temperature that flexible substrates can withstand during the fabrication process for flexible electronics.

In this work, we fabricated oxide TFTs with IGZO channel layers on flexible substrates. To examine the importance of the IGZO channel composition and provide a suitable composition for flexible device applications, the device

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characteristics were compared when the IGZO composition was 2:1:2 vs 2:2:1 (In:Ga:Zn). We successfully confirmed that the device stability and reliability of the fabricated IGZO (2:1:2) TFT were quite suitable for flexible electronics applications.

II. EXPERIMENT

We fabricated IGZO TFTs with a top-gate bottom-contact structure. A polyethylene naphthalate (PEN) film with a thickness of 125 μm was used as the flexible plastic substrate. For the first step, the PEN films were laminated on a glass substrate using cool-off type adhesive (Intelimer®) by Nitta Corp. (Japan) for convenience of device fabrication. This process step served the critical role of delaminating the PEN film from the carrier glass substrate after terminating all processes. The strength of the employed adhesive could be controlled by adjusting the temperature. Then, the Dongjin organic insulator (manufactured by Dongjin Semichem) and Al_2O_3 layers (serving as organic and oxide buffer layers, respectively) were prepared on the laminated film to yield a substrate with a smooth surface, low moisture permeability, and strong protection from mechanical damage. Thick (200 nm) Ti electrodes were subsequently deposited via sputtering as source/drain electrodes. The sheet resistance of the Ti layer, as measured by the four-point probe method, was approximately 6 Ω/□. The 20 nm-thick IGZO channel layers were deposited by dc sputtering at room temperature. The IGZO channels were made with compositions of 2:1:2 and 2:2:1 (In:Ga:Zn). In order to prevent chemical damage and environmental effects to the channel layers, 9 nm-thick Al_2O_3 layers were formed by atomic layer deposition (ALD) at 150 °C.[27] After a one-step patterning process to define the active area of Al_2O_3/IGZO, a 176 nm-thick Al_2O_3 gate insulator layer was grown via ALD at 150 °C using trimethylaluminum and H_2O (as Al and oxygen precursors, respectively). Finally, a 200 nm-thick Ti layer was deposited by rf sputtering to serve as a gate electrode after the formation of contact holes, which establish electrical contacts for the devices. The deposition rates for all layers and the corresponding film thicknesses were measured by a surface profiler. All patterning processes for device fabrication were performed using conventional photolithography and wet etching with a diluted hydrofluoric acid-based etchant.

A final heat-treatment was performed at 150 °C for 2 h in a vacuum below 10^{-2} Torr, as is conventionally prescribed for the optimization of device performance. The annealing temperature (150 °C in this work) was chosen according to the thermal stability of the PEN film.

For the final step, the laminated PEN films on which the IGZO TFTs were fabricated were carefully detached from the carrier glass substrate by decreasing the temperature (below 10 °C).[28] A schematic diagram of a fabricated flexible device is shown in Fig. 1. The electrical characteristics of the fabricated devices were measured with a semiconductor parameter analyzer (Agilent, B1500 A) in a dark box. The defined gate channel width (W) and length (L) of the evaluated devices were 40 and 20 μm, respectively.

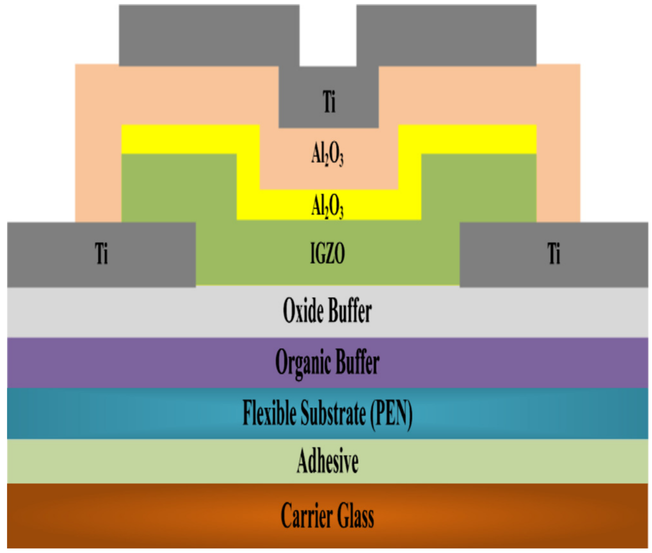

FIG. 1. (Color online) Schematic cross-sectional diagram of a fabricated IGZO TFT on a flexible PEN substrate.

III. RESULTS AND DISCUSSION

Figure 2(a) shows the mask align patterns for the process for IGZO TFTs fabricated on a laminated PEN film. A design rule of 10 μm is used. As evident in the figure, the lamination process performed in this work was fully compatible with the conventional lithography process, even on the plastic PEN film. Photographs of both a PEN film laminated onto the carrier glass before device fabrication and delaminated flexible devices after the full fabrication process are shown in Figs. 2(b) and 2(c), respectively. No air bubbles were found between the laminated film and the carrier glass or adhesive. For the delaminated film, we also found no mechanical damage. These observations suitably reflect the integrity of our fabrication process for producing flexible IGZO TFTs.

The transfer characteristics of IGZO TFTs with the varying compositions of 2:1:2 (212-IGZO) and 2:2:1 (221-IGZO) are displayed in Figs. 3(a) and 3(b), respectively. The 212-IGZO device exhibited excellent electrical characteristics even though it was treated at a relatively low annealing temperature of 150 °C. The μ_{FE}, V_{TH}, and S.S were calculated to be approximately 7.83 cm V^{-1} s^{-1}, 1.93 V, and 0.24 V/decade, respectively. The V_{TH} value was determined by adjusting the gate voltage, which induces a drain current of L/W × 10 nA at a V_{DS} of 15.5 V. In addition, the S.S (V/decade) was extracted from the linear portion of the log (I_{DS}) versus V_{GS} plot. The μ_{FE} was calculated from the following equation:

$$I_{DS} = \frac{W}{2L}\mu_{sat}C_{ox}(V_{GS}-V_{TH})^2,$$

where C_{ox} is the capacitance of the Al_2O_3 gate insulator. On the other hand, the 221-IGZO device did not show transfer characteristics at an annealing temperature of 200 °C, as shown in Fig. 3(b). Considering the thermal budget for the PEN film is limited to not more than 200 °C, the use of 221-IGZO in flexible electronic applications does not seem

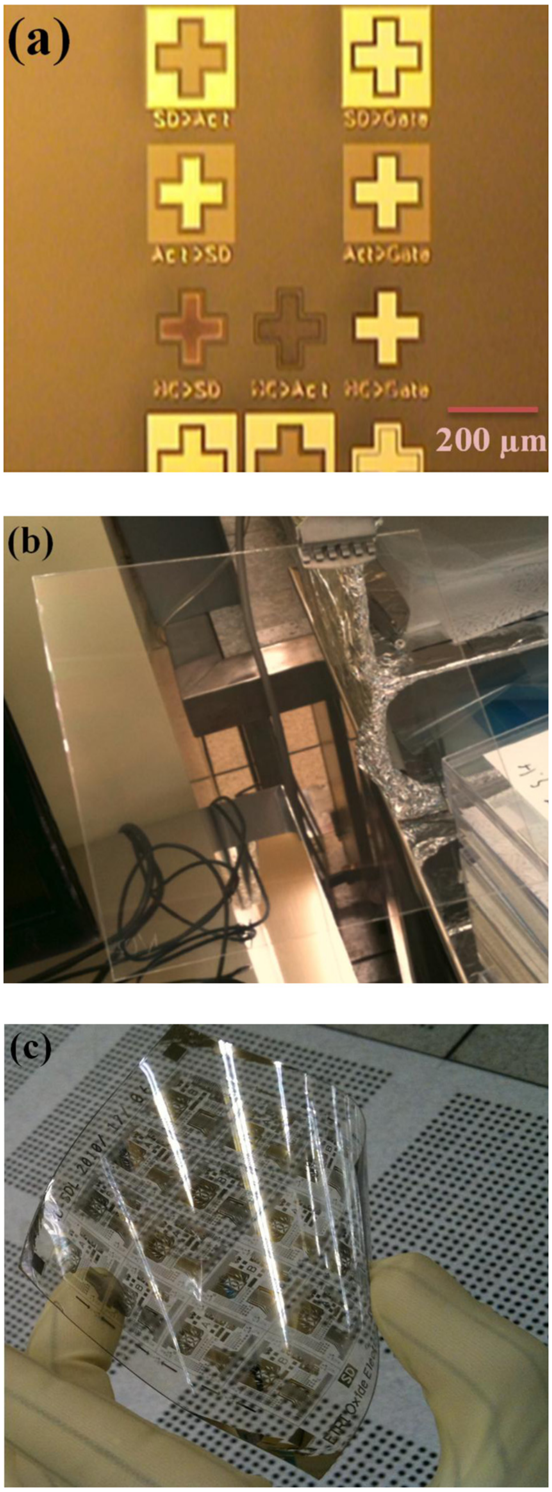

FIG. 2. (Color online) (a) Mask align patterns of the fabricated flexible devices and photographs of (b) a PEN film laminated onto a carrier glass substrate and (c) the flexible devices after the delamination process.

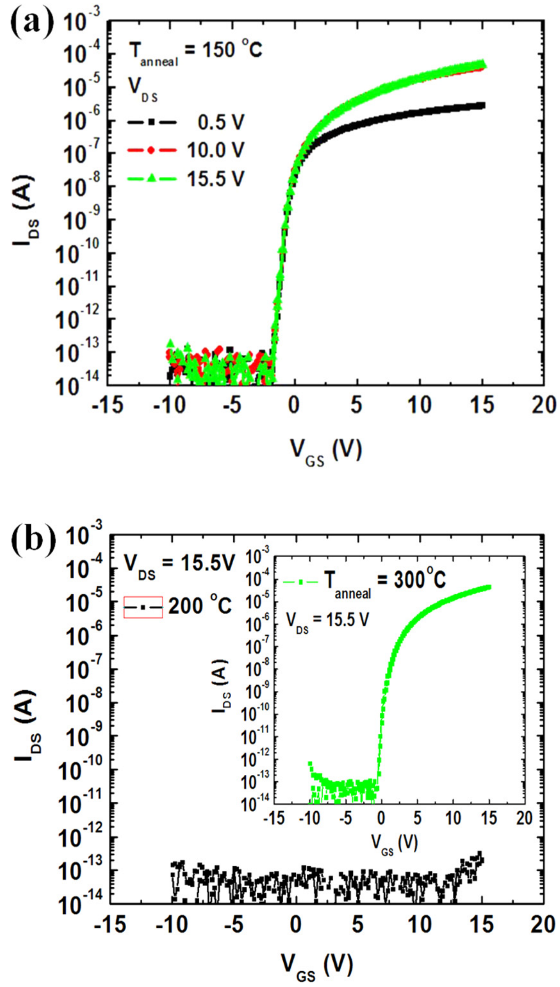

FIG. 3. (Color online) I_{DS}-V_{GS} transfer characteristics (in logarithmic scale) of (a) the 212- and (b) the 221-IGZO TFTs fabricated on a PEN substrate. The final annealing temperatures of the devices in (a) and (b) were 150 °C and 200 °C, respectively. The inset of Fig. 2(b) shows the I_{DS}-V_{GS} transfer characteristics of 221-IGZO TFTs fabricated on a glass substrate and annealed at 300 °C.

reasonable. To confirm the feasible temperature for the 221-IGZO to optimize its device performance, we also prepared a 221-IGZO device on a conventional glass substrate as a control device; the device structure and fabrication processes were exactly the same as those for the PEN substrate. As shown in the inset of Fig. 3(b), normal operation of the 221-IGZO device could not be confirmed until the annealing temperature was increased to 300 °C. The μ_{FE}, V_{TH}, and S.S for this control device were 9.52 cm V^{-1} s^{-1}, 3.23 V, and 0.26 V/decade, respectively.

It was evident from the stated results that the optimum annealing temperature for the IGZO TFTs was highly dependent on the composition of the IGZO. Although the 212-IGZO has almost the same In composition as 221-IGZO, the optimum annealing temperature for the 212-IGZO device could be decreased by 150 °C when compared to that of the 221-IGZO TFT. From the viewpoint of device characterization, it is generally accepted that a high In content incorporated into the oxide active channel can enhance the carrier mobility and shift the turn-on voltage in a negative direction.[16] Actually, for the case of a Hf-In-Zn-O channel for TFT applications, the amount of Hf may also be an important factor in determining the required temperature range to optimize the channel properties for TFT operations.[15] However, given the device parameters obtained in this work, we cannot simply say that only the In composition has an important effect on the overall electrical performance when considering the required thermal budget. In other words, these behaviors could be caused by complex interactions among the constituent In-Ga-Zn compositions. Therefore, the reason why the 221-IGZO device did not work when annealing was conducted below 300 °C and why the 212-IGZO device showed better device performance even at a lower process temperature may be related to the difference in activation energy for the rearrangement of the constituent elements in the channel layer among different IGZO compositions, which plays an important role in determining the required process temperature for the TFTs. Once the IGZO film is activated by an appropriate thermal treatment, the generated carriers within the IGZO film with a given composition can have a dominant effect on the device characteristics. While it is very challenging to accurately analyze this issue because critical evidence is very difficult to obtain for amorphous thin films, we suppose that the rearrangement of atoms is a crucial process for obtaining an appropriate carrier concentration and channel conductivity for the oxide active layer. This process requires some activation energy that obeys a typical Arrhenius relationship.

To confirm the operational stability and process reliability of the fabricated 212-IGZO device, transfer curves of the TFTs were obtained, as the channel lengths were varied and the channel width was held constant at 20 μm; the results are displayed in Fig. 4(a). All of the transfer characteristics were observed to be very stable regardless of the patterned size of the active area. Figure 4(b) shows the output characteristics of the 212-IGZO device with a W/L of 40/20 μm when the V_{GS} was set to 0, 2, 4, 6, 8, and 10 V. It was confirmed that the output curves were very stable without current crowding. It is also very important to examine the bias-stress stability of the flexible IGZO TFTs. Figures 5(a) and 5(b) show the operational stabilities of the 212-IGZO device under positive or negative gate bias stress conditions, respectively, in which a V_{GS} of 20 V or −20 V was applied as a bias stress for 10^4 s in a dark box. As evident in Fig. 5, the transfer characteristics remained highly stable despite a relatively low annealing temperature of 150 °C. In addition, no noticeable change in the transfer characteristics was observed. For oxide TFTs, stability characteristics are dependent on environmental conditions in which oxygen and/or water molecules exist. The introduction of a passivation layer can be very effective to protect the device from these environmental effects.[18,29]

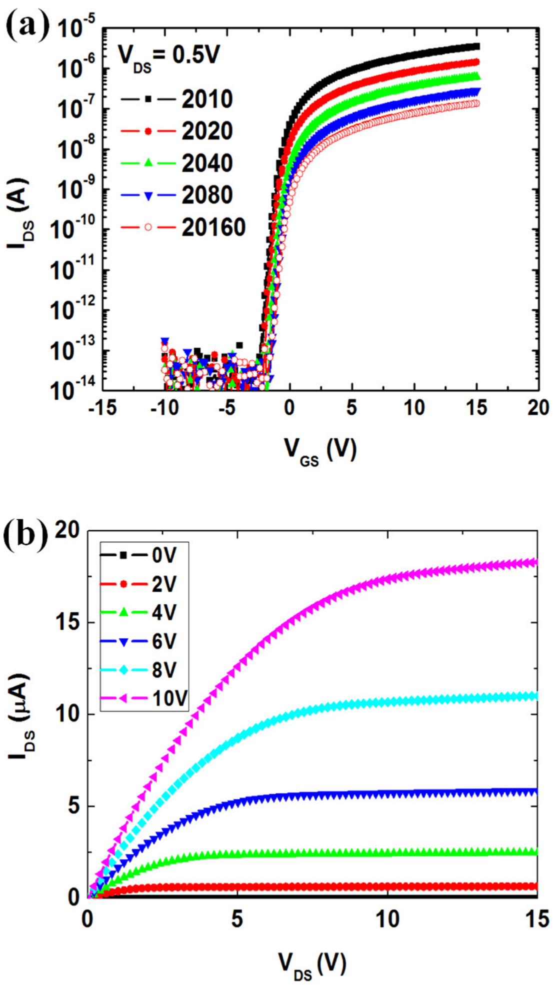

FIG. 4. (Color online) (a) I_{DS}-V_{GS} transfer characteristics of the flexible 212-IGZO TFTs when the channel length was varied from 10–160 μm. The channel width of all devices was fixed at 20 μm. (b) I_{DS}-V_{DS} output characteristics of a flexible 212-IGZO TFT with a W/L ratio of 40/20 μm.

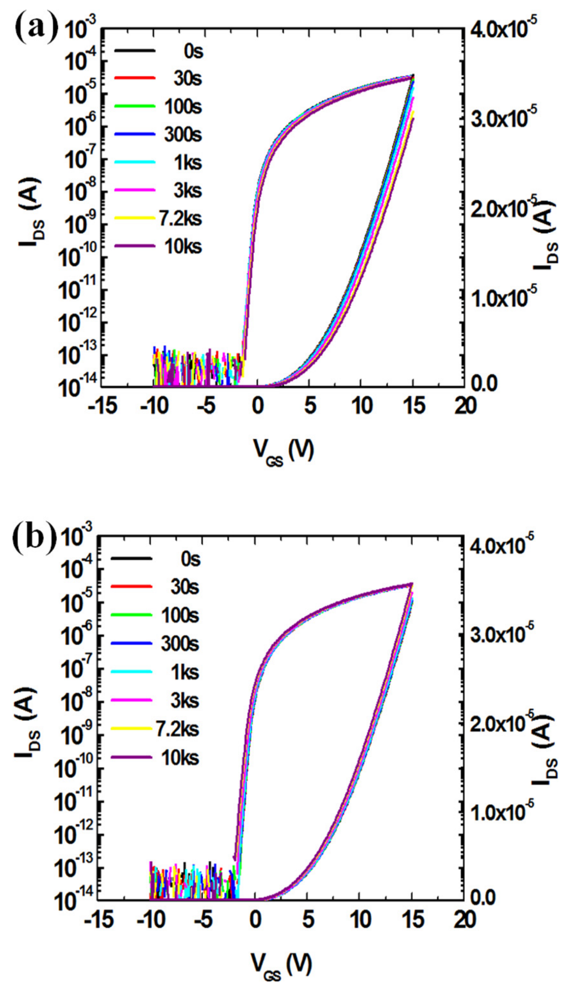

FIG. 5. (Color online) Transfer characteristics of flexible 212-IGZO TFTs under a gate bias (V_{GS}) of (a) 20 and (b) −20 V. Stabilities against the bias stress were examined for 10^4 s.

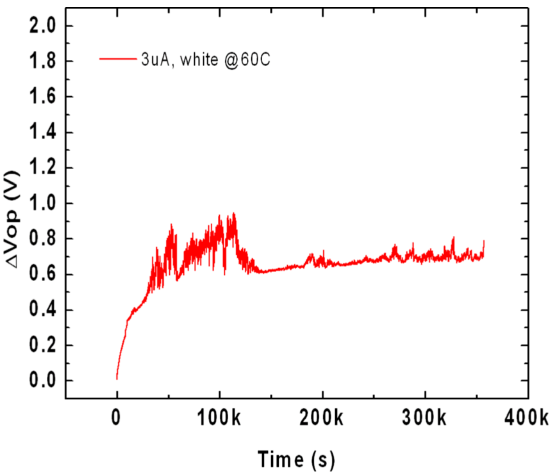

FIG. 6. (Color online) Variations in the operation voltage of the flexible 212-IGZO TFTs under a constant current stress (3 μA) with light illumination (white—1 mW/cm^2) at 60 °C. Stabilities against the bias temperature light stress were examined for up to 3.5×10^5 s.

The excellent stability characteristics of the flexible IGZO TFTs may be due to the dense ALD-grown Al_2O_3 layer.

Finally, we investigated the stability of the 212-IGZO device under a constant current stress with light illumination at a temperature of 60 °C. The drain current flowing across the device was maintained at 4 μA for 3.5×10^5 s. The power of the irradiated visible white light was 1 mW/cm^2. Variation in the operation voltage (V_{OP}) is defined as the voltage where the I_{DS} approaches 1 pA from the off state in the transfer characteristics, and is shown in Fig. 6. Despite the rigorous stress test conditions that are typically employed for evaluating back-plane devices for active-matrix organic light emitting diode displays, the 212-IGZO device showed great stability with low V_{OP} variations ($\triangle V_{OP} \leq 0.9$ V).

The device stability before/after the lamination process under bending conditions is also very important for future flexible electronics applications. These characteristics will be investigated and discussed in detail in our future studies.

IV. CONCLUSIONS

IGZO-channel oxide TFTs were fabricated on flexible PEN substrates. Two strategies were proposed to realize high-performance flexible IGZO TFTs at a lower process thermal budget than conventional strategies allow lamination and delamination processes were well established to facilitate the fabrication process for flexible IGZO TFTs. Then, a 2:1:2 (In:Ga:Zn) IGZO channel composition was proposed. The final annealing temperature requirement was found to be significantly reduced from 300 °C to 150 °C when the IGZO composition was changed from 2:2:1 to 2:1:2. It can be concluded that the optimum process temperature was strongly dependent on the complex effect of the In-Ga-Zn compositional ratio rather than the absolute In content within the IGZO channel layer. This was confirmed by variations in the activation energy for the rearrangement of the constituent elements in the channel layer at different IGZO compositions. For the successfully fabricated IGZO TFT with a composition of 2:1:2, μ_{FE}, V_{TH}, and S.S were found to be approximately 7.83 cm^2 V^{-1} s^{-1}, 1.93 V, and 0.24 V/decade, respectively. It was also confirmed that the fabricated flexible IGZO TFT exhibited excellent device stability characteristics under a positive/negative gate bias-stress and illuminated constant current stress conditions. From these results, we conclude that IGZO-channel oxide TFTs fabricated using our methodology can be promising device components for highly functional flexible electronics applications.

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