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Role of Composition, Growth Technique, Temperature, Substrate Effects on the Efficiency of an IGZO Thin Film Transistor

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Abstract:Currently,Metal oxide semiconductors have become popular as a suitable active-layer for thin-film transistors.Due toitsimportance in display technology. Commonly, displays rely on TFTs of amorphous hydrogenated silicon (a-Si:H), but the needis for large area displays with higher resolution, fast response time, lower power consumption and compatibility with integrated driving circuits have prompted research into other semiconducting materials. As a result, metal oxides have become major prospects to replace a-Si:H with their high-performance electrical characteristics and simplicity of processing, making them valuable switching elements in display technology. Particularly, quaternary metal oxide semiconductors such as the a-IGZO have discussed extremely high performances as TFTs, prompting extensive research in the field.In solution-processed IGZO, there have been a couple approaches to improve device performance and stability as well as simplify processing. In this work, we produce a gallium-rich 2:2:1 IGZO TFT using solution processes and study its electrical characteristics and stability. In this paper, we demonstrate a working solution processed gallium-rich 2:2:1 IGZO TFT and compare it to a solution-processed indium-rich device to quantify its stability and performance.

Keywords:zinc oxide semiconductor, flexible and transparent electronics, oxide TFT's , IGZO TFT.

I. INTRODUCTION

In current years, the oxide-based semiconductors are being widely investigated for their increased mobility, greater transparency, lower processing temperature and good uniformity[1-10]. Displaymanufacturers began to pay attention to oxidesemiconductors because manufacturing is simpler less complicatedwith that of crystallized-silicon TFTs and can reduce the maximum process temperature,allowing for use in large-sized displays and substrates. In display technology, these TFTs arehighly sought after as switches for their switching components for wideareas, ultra-high definitionand fast frame rates to turn pixels on and off. There are no grain boundaries for Amorphous oxide semiconductors (AOSs) and they are restricted notmuch like polycrystalline semiconductors.

As such, amorphous metal oxide TFTs are currently strong candidates for AMOLED backplanes. With increasing performance demands in flat-panel displays especiallyimproved resolution and frame rate exceeding the limits of a-Si:H TFTs performance, high performance TFTs will also be needed for the future AMLCD where oxide TFTs could potentially replace a-Si:H as well as replacing LTPS in AMOLED. Specifically, in the past decade an IGZO

TFT revealedverysuitablechoicefor flexible displays and AMOLED TV because of its higher mobility of 10-40 $cm^2/(V\cdot s)$, amorphous structure, and substantiallylower thermal budget processes [12,13].

II. Metal Oxides for Thin Film Transistors

SinceTFTs are invented, many materials have been used as active layers. Mostprominently, a-Si:H has been the most common material employed for TFT active layers. One material that has garnered significant interest in recent years is the metal oxide semiconductor for TFT applications, particularly in the amorphous state [17].

Oxide semiconductor materials have increasingly shown attention in recent past years mainly due to the reasons of two characteristics: optical transparency and electrical conductivity. ITO has been commonly used as a transparent electrode material for solar cells and displays because of its good transparency and conductivity. Early on, most attention on oxides was been limited to transparency and conductivity, while its potential as a functional material forsemiconductor devices was still being considered.

Nomura and Hosono first released a high-performance monocrystal transparent oxide transistor using IGZO, later followed by a TFT deposited at room temperature with amorphous IGZO [8, 9, 17].The first work demonstrated the high-performances that could be found in metal oxide semiconductors while his second work demonstrated their potential with low-temperature fabrication. In spite of the room temperature deposition, the amorphous device exhibited high mobility, ten times that of amorphous silicon. Which has since made oxide semiconductor materials a subject of increasing attention as an active layer for display-use TFTs. With mobilities compared to crystalline oxides semiconductors and large energy band gaps allowing for transparency in the visible spectral range, metal-oxide semiconductor (MOS) TFTs have emerged in electronic device markets as new applications such as intelligent wearable systems[19, 20], epidermal devices[21, 22], artificial skin[23, 24], medical implants[25, 26], liquid crystal displays[27] , transparent transistors[15, 16, 28], transparent oxide memory [29, 30], solar cells [31, 32], electrophoretic displays, electrochromic windows, electro-optical devices, paper electronics [33, 34], and gas sensors[35-37]. Similar to conventional MOSFET devices, MOS TFTs can also be used in integrated circuit designs, such as line drivers for AMOLED, logical circuits, digital-to-analog converters, RFID or NFC applications [38-41].

Table 1. Differences between silicon and metal-oxide TFTs in electrical characteristics and processing.[42]

Material	**Amorphous Si**	**Low Temperature Poly-Si**	**Metal Oxide Semiconductor**
Carrier Mobility	< $1cm^2V^{-1}s^{-1}$	$50\text{-}100cm^2V^{-1}s^{-1}$	$10\text{-}100cm^2V^{-1}s^{-1}$
Subthreshold Swing	0.4-0.5V/dec	0.2-0.3V/dec	0.09-0.6V/dec
Leakage Current	10-12A	10-12A	10-13A
Uniformity	Good	Poor	Good
Number of Masks	4-5	5-9	4-5
Manufacturing Cost	low	high	Low
Process Temperature	≈250°C	≈250°C	Room temperature to ≈350°C

Metaloxide semiconductor TFTs shown much prominence on device properties compared to conventional technology such as lower manufacturing cost, high scalability, low process complexity and temperature as shown in Table 1[42].

These amorphous oxide semiconductors (AOSs) exhibit potential with their combination of higher optical transparency, large electron mobility and amorphous [43,44] microstructure. Lacking grain boundaries, AOSs doesn't face theprimary limitation of mobility in polycrystalline semiconductors, creating a huge advantage inprocess integration. Other advantages include low temperature deposition routes and ultra-smooth surfaces that suppress interface traps and scattering centers. These TFTs can also be fabricated on flexible-substrates while maintaining high performances, increasingtheir potential applications. In addition, the materials used in this technology are eco-friendly and less expensive than existing technologies [11].

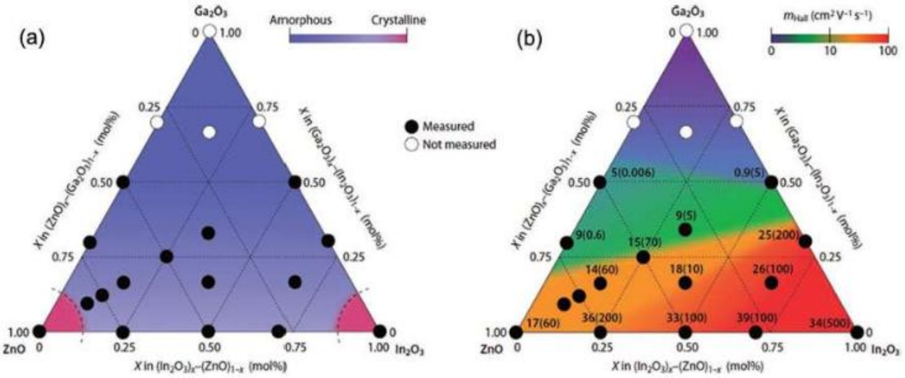

Figure 1. (a) Crystalline and (b) Hall-effect mobility of IGZO metal-oxide semiconductors with different atomic ratios.[45]

In Figure 1, there are metal composition triangles for indium, gallium, and zinc ratios that show (a) crystallinity and (b) mobility findings and trends for specific composition ratios.

Currently, there are many successful metal oxide semiconductors being used in different applications. ZnO[14, 47, 48], IGZO [15, 49], ZIO [50, 51], and ZTO [52, 53] are a couple of examples of popular metal oxides because of their wideband gaps, very high transparency, and outstanding electrical properties. Moreover, zinc-oxidebased transparent-semiconductors have been found to be applicable to sol-gel process, which allows low-cost and printable production [54-58]. Among these, due to its high channel mobility, high on-off ratio, low toxicity, excellent environmental stability and good optical transmittance, IGZO is a main semiconductor for transparent oxides TFTs, which has led to its application in many display technologies [59].

As such Ga is commonly used as a suppressing cation. IGZO was a breakthrough in metal oxide research.As themember of AOS, IGZO having high mobility of 10-30cm^2/(V·s), good stability, lower process complexity, and already been produced commercially in the display industry.

III. Composition Engineering of IGZO

Solution-processing of IGZO and its importance, followed by studies on the effects of the metal composition ratio of IGZO. By establishing the background information for these topics, we can demonstrate the importance of our contribution to the field: applying solution-process to fabricate a gallium-rich 2:2:1 IGZO TFT and evaluating its performance and stability.

IV. Solution-Processing Advantages

Solution-processed oxide TFT’s usesdipping, spin-coatingand inkjet printing. They have attracted significant attention due to vacuum deposition processes such as RF magnetron sputtering and pulsed laser deposition require complicated and high manufacturing cost processes. Due to its low cost and lower temperature processing nature, solution-process based oxide TFTs have several advantages and they are ideal for large area, high-throughput and flexible displays [46].

Additionally, they won’t require sophisticated vacuum systems or costly equipment typically found in vacuum deposition methods [60, 63]. Another advantage of solution process deposition is avoiding environmentally harmful templates which are mostly used for the synthetics of inorganic materials use in vacuum deposition methods. Even though the mobilities of the solution-based deposition method devices are lesser-than their vacuum deposition counterparts due to the fine control of vacuum processes.The chemical solution deposition method shows great importance in the fabrication of amorphous TFTs [64].

For IGZO, solution-processing is ideal, particularly for display technology, where the amorphous thin film can be applied over large areas simply and cheaply. There have been many cases of successful solution-processed IGZO TFTs, such as Yoon's IGZO of 5:2:1 ratio [65].

While many compositions have been investigated for solution-processing and found that certain compositions did not favour amorphous structure, which can lead to poor electrical performances [18].

V. Metal Composition Effects

Among the metal oxides, amorphous IGZO wasamong the most attractive materials forTFTs because ofuniform characteristics, higher field effect mobilityand compatibility with lower temperature processed [8,66].Barquinha, reported on the effects of the processing parameters such as target composition using several fixed compositional IGZO targets on the electrical properties of IGZO-TFTs [44]. Kim, investigated the combinatorial effects on surface morphology andelectrical performance of a-IGZO TFTs in solution-processed IGZO [18] and Lee, investigated electrical-characteristics of solution-processed IGZO TFTs with different composition ratios of precursors (7:1:2,6:3:1,5:1:4) and analysed the effects of composition ratioswith solution-processedIGZO-TFTs[70].

Kim’s Hall measurements with varying Inand Ga molar ratios (8:1:1, 8:1:8, 8:3:5, 8:8:5, 4:1:5, 5:1:5, 8:1:5) found that increasing In increased the average carrier concentration and Hall mobility in the IGZO devices.Where the increased drain current and carrier concentrations could be explained by enlarged In 5s orbital at the bottom of the conduction band from additional In3+ ion incorporation in the structure [18,54,67]. Kim, also found that a high atomic concentration of In >80% resulted in polycrystallineIGZO films [55, 60, 61, 71].Whereas with lower In content, the films stayed in the amorphous phase. Also, a high atomic concentration of In or Zn>80% produced films with nonuniform surface morphology>2.5nm and a large grain size>100nm, which would make inconsistent and poor-performing devices.

In Barquinha’s work, they also found that their indium-rich composition of 4:1:2 led to a highly conductive film and uncontrollable channel conductivity inthe devices [44].

Zinc composition was found to influence subthreshold characteristics. Barquinha's best device results came from a 2:1:2 device, where the increased zinc content (compared to 2:1:1) allowed for enhanced switching behaviour and lower SS. The increased Zn content reduced conductivity and mobility from the 2:1:1 device but allowed for better switching through SS decrease [44]. Zn content in the film is believed to cause change in the subthreshold characteristics by modulating shallow tail states or by reducing the interfacial states at thechannel/gate insulator interface [62]. Lee et al. found that with an increase in Zn content to 5:1:4, device hysteresis decreased because Zn modulated the shallow tail state below the conduction band and reduced the interstitial states between channel and insulator [70].

For the composition studies, gallium appears to have the opposite effect of indium, where increased gallium content results in reduced carrier concentration and mobility. Kim, found that the critical decrease of Hall mobility and carrier concentration were observed by increasing Ga content, which they attributed to possible Ga substitution of Zn interstitials or suppression ofoxygen vacancy formation [62]. They found that Ga content is also effective in controlling V_{ON} and drain current by increasing electronegativity of the films or by reducing carrier concentration. When Ga composition increased in Lee et al.'s 6:3:1 IGZO, the off current decreased because Ga compensated carriers generated by In conclusion and suppressed formation of oxygen vacancies. Gallium-rich devices decreased off current by suppressing formation of oxygen vacancies [70]. Lastly, Barquinha, found that increased gallium content in their IGZO resulted in good switching devices with low V_{TH} and SS, with significantly lower mobility [44]. However, they found that increasing gallium content too much resulted in a highly resistive films and poor devices with high SS and V_{TH}. Barquinha's results indicated that the IGZO electrical properties are sensitive to gallium content, where too much would cause asignificant reduction in the device characteristics. The proper amount of gallium allowed for lower V_{TH} and SS which would benefit stability in these TFT devices.

In addition to compositional studies on IGZO electrical performances, researchers such as Huh[68] and Cheong [69] have also studied compositional effects upon IGZO device stability.

The compositions compared were 2:1:2 (device A) and 2:2:1 (device B) of In:Ga:Zn. Device A exhibited larger driving current and greater field effectmobility than device B but it had a more negative turn-on voltage at -4.8V versus -0.8V. Thecarrier density in IGZO A TFT was also higher at a gate voltage of 0V than device B. What Huhet al. found was that as the Ga content exceeds the Zn content, the turn-on characteristics of TFTdegraded, but the stability enhanced. Where, IGZO device B with higher Ga content showed lesserfield effect mobility. The subthreshold slopes of the two devices were nearly the same at 0.35V/dec versus 0.39V/dec and Huh calculated the densities of interface trap states: IGZO A had 1.00×1012cm^{-2} and IGZO B had 1.14×1012cm^{-2}[68]. The difference in field effect mobilityof 14$cm^2/(V\cdot s)$ and 9.3$cm^2/(V\cdot s)$ can be attributed to the bulk characteristics.

The effect of composition on device stability was also investigated, showing the changes in transfer characteristics before and after electrical bias stress using a positive gate bias of 20V continuously applied for 10000 seconds at a substrate temperature of 340 K. The heat was applied to accelerate device degradation and IGZO B with higher Ga content showed more stable characteristics. Overall, Huh found that the composition of device A 2:1:2 had a superior mobility by 5$cm^2/(V\cdot s)$, but had a poor turn-on voltage at -4.8V, much worse than the device B2:2:1, which yields a turn-on voltage of -0.8V[68]. While the mobility of device A was found to be superior, both devices exhibited acceptable and good mobilities. However, device A's mobility could not compensate for its poor turn-on voltage and poor stability, which rendered it unusable as a device. Device B's superior turn-on voltage and stability was a worthy trade-off for a small drop in mobility making it a more highly valued candidate for TFT applications.

Cheong reports three sputtered IGZO compositions used: 2:1:2, 1:1:1, and 2:2:1. Cheong studied electrical instability of IGZO-TFTs with varying compositions. According to his tests, the larger Ga content, the more robust the TFT, consistent with x-ray photoelectron spectroscopy analysis.The strong binding of Oatoms appeared in the IGZO film with larger Ga[69]. The 2:1:2 device with low gallium had a V_{ON} of -1.26V, mobility of 18.09cm^2/(V·s) and SS of 0.20V/dec. The 1:1:1 device had the lowest mobility of 7.9cm^2/(V·s) with V_{ON} of 0.21V and SS of 0.26. Finally, the 2:2:1 device with high gallium had a V_{ON} of 0.42V, mobility of 9.12cm^2/(V·s) and SS of 0.29V/dec[69]. These results show that mobility does decrease significantly for the devices with higher gallium content while subthreshold swing does not change by much. Most importantly, the V_{ON} for the devices with more gallium were much closer to 0V, indicating strong candidates for better devices. Under positive bias stress (V_G = 20V), over 100000seconds, the 2:1:2 V_{TH} shifted by 1.7V, the 1:1:1 device V_{TH} shifted by 0.5V, while the 2:2:1 device V_{TH} shifted by -0.2V. Similar results were obtained through constant current stress tests, where the high gallium 2:2:1 device noticeably outperformed the lower gallium devices in stability. From this data and stress tests it can be concluded that the higher gallium content 2:2:1 device had a smaller absolute shift in threshold voltage than the lower gallium devices,outperforming the low gallium 2:1:2 device significantly. Such a result while preserving a good mobility of 9.12cm^2/(V·s) show that the increased gallium content in this device’s composition does not hinder the mobility significantly while improving stability several times over.

Indium content correlated with increased mobility and off-current as the In3+ ion has been reported to form extended conduction band minima by the percolation of In 5s orbitals and provide the main electron conduction path [8]. However, thedownside of too much indium was a device that could not be controlled and turned off. Kim et al. also found that too much indium content resulted in polycrystalline films. Zinc content has been found to induce changes in the subthreshold characteristics thought to be done by modulating shallow tail states or reducing interfacial states at channel/gate insulator interface. Finally,gallium was found to decrease mobility and carrier concentration, due to the superior oxygen binding of Ga3+ ion which allows for suppression of free carriers from oxygen vacancies as Ga content increases [10, 8]. Since Ga-O bonding is much stronger than either Zn-O or In-O bonding, it is more challenging to generate charge carriers through the creation of oxygen vacancies. While gallium decreased the mobility, it was found to favour a V_{TH} or V_{ON} much closer to 0V and devices withhigh stability.

With these results in mind, devices can be carefully designed to meet specific requirements. For our interest in high stability, gallium content is of prime importance, as work done by Huh and Cheong have successfully demonstrated higher stability in devices with increased gallium content. In Barquinha’s work, the mobility decrease in a gallium-rich device was considered almost negligible, making it a high-performance device [44]. With too much of gallium however, the device characteristics degrade, demonstrating that there is likelyan optimal concentration to maximize performance and stability.

VI. Synthesis of Metal-Oxide Solutions:

The 0.1M IGZO solution was synthesized by dissolving indium nitrate hydrate ($In(NO_3)_3·xH_2O$, Aldrich, 99.999%), gallium nitrate hydrate ($Ga(NO_3)_3·xH_2O$, Aldrich, 99.999%) and zincacetate dihydrate ($Zn(CH_3COO)_2·2H_2O$, Aldrich, 98%) powders in 2-methoxyethanol (2ME,Aldrich, 99%) in a total metal ion molarity of 0.1 M.The molar ratios of the fabricated devices were 9:1:2 and 2:2:1 of In, Ga, and Zn for the twoIGZO devices.

We fabricated devices using precursors to produce IGZO devices of 9:1:2 and 2:2:1 metalcomposition ratio. From the transfer curves we were able to get the device electricalcharacteristics listed Table 1 below.

Table 1: Electrical parameters (μ, V_{TH}, on/off, SS) of 2:2:1IGZO and 9:1:2 IGZO devices compared side by side

Electrical Parameters	**2:2:1 IGZO**	**9:1:2 IGZO**
Mobilityμ	1.1 cm^2/V.s	15.9cm^2/V.s
Threshold-voltageV_{TH}	-0.17V	14.11V
On/Off-ratio	$3.52x10^7$	$1.20x10^7$
Subthreshold Swing	0.75V/decade	1.48V/decade

Overall, the 2:2:1 IGZO experienced a much smaller shift in turn-on and threshold voltage, while the 9:1:2 IGZO shifted dramatically. The 2:2:1 voltage shifts were on a much smaller scale, with shifts less than 1V. In contrast, the 9:1:2 device shifts typically got worse as longer stress was applied, shifting over 10 V by the end of the stress tests. The 9:1:2 device experienced a shift almost 25-40 times the shift of the 2:2:1 device.

The 9:1:2 device shows great mobility of 15.9 $cm^2/(V\cdot s)$, while the 2:2:1 device has a mobility of 1.1$cm^2/(V\cdot s)$. Although the 9:1:2 device holds the advantage in mobility, the other electrical characteristics favour the 2:2:1 device: a significantly better V_{TH} (-0.17V vs 14.11V), SS (0.75V/dec vs. 1.48V/dec), and slightly better on/off ratio (3.52×10^7 vs 1.20×10^7). Just based on these results, solution-processed 2:2:1 IGZO is a viable material for TFT technology. It does notpossess great mobility but still outperforms a-Si:H. More importantly, it possesses great V_{TH} close to 0V, SS less than 1V/dec and have a strong on/off-ratio higher than 10^7. With these characteristics, the gallium-rich 2:2:1 IGZO is not only a functional device but also a strong candidate for devices.

The 2:2:1 device is superior to the 9:1:2 device in stability. The indium-rich 9:1:2 IGZO composition shifted significantly after each stress test, while the 2:2:1 experienced little to no shift.

Table 2: change in Threshold voltageand turn-on voltage at each time point following electrical bias stress for both 2:2:1 IGZO and 9:1:2 IGZO devices.

Time	**2:2:1 IGZO ΔV_{TH}**	**9:1:2 IGZO ΔV_{TH}**	**2:2:1 IGZO ΔV_{ON}**	**9:1:2 IGZO ΔV_{ON}**
0s	0V	0V	0V	0V
100s	0.483V	1.142V	0.4V	0.7V
200s	0.535V	1.945V	0.4V	1.4V
500s	0.572V	4.024V	0.4V	3.5V
1000s	0.600V	6.340V	0.4V	6.3V

2000s	0.634V	9.182V	0.4V	10.5V
5000s	0.635V	13.630V	0.4V	15.4V
10000s	0.638V	15.004V	0.4V	18.2V

From Table 2 and Figures 2 and 3, it can be observed that the $\Delta V_{TH}/\Delta V_{ON}$ for the 2:2:1 IGZO was approximately 0.4-0.6V throughout the entire stress test. In contrast, the 9:1:2 IGZO experienced shifts of 15-18V in the same stress time, a difference in stability of approximately 25-40 times. This huge disparity in stability shows that the 9:1:2, despite its high mobility, lacks stability, while the 2:2:1 is impressively stable under the same conditions.

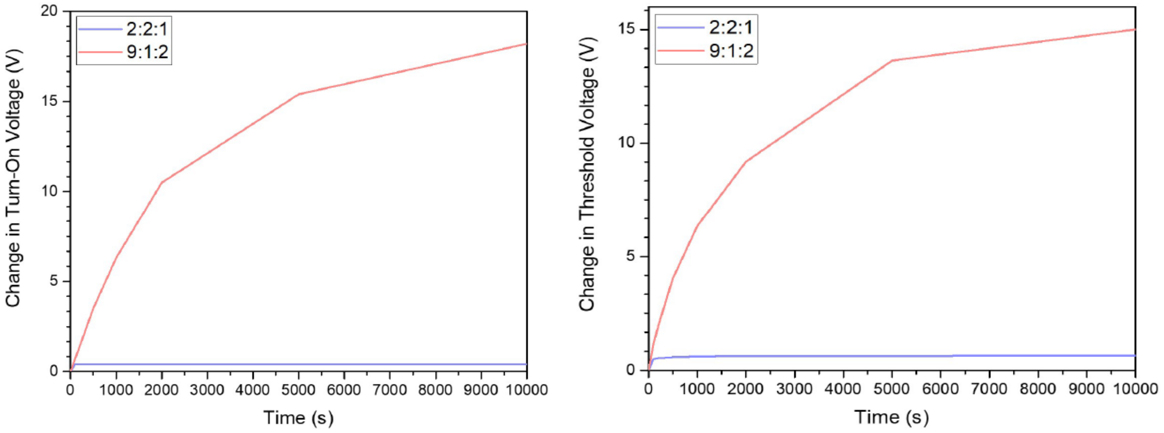

Figure 2 (left) and 3 (right). Shift in turn-on voltage (left) and threshold voltage (right) over time for both 2:2:1 IGZO and 9:1:2 IGZO devices.

The electrical characteristics can be traced back to composition. The high indium content in the 9:1:2 is likely responsible for the impressive mobility. The indium content is not excessive, as the IGZO TFT still displays normal transfer curves with currents that can be controlled on and off. In the 2:2:1 IGZO, the gallium content increases from 8% to 40% and Ga- O bonds are very effective in reducing oxygen vacancies in the TFTs, making the turn-on and threshold voltages close to 0V. Under the stress tests, the 9:1:2 is highly unstable, likely because the number of oxygen vacancies and high-carrier concentration which increase its variability. In contrast, the 2:2:1 IGZO which has reduced vacancies and carrier concentration does not shift as much and is more stable. These results demonstrate that the gallium-rich 2:2:1 IGZO TFT we fabricated through solution-processing is a functional device with good electrical characteristics.

On top of this, through comparison with the indium-rich 9:1:2 IGZO device, the solution processed 2:2:1 IGZO demonstrates amazing stability. In this way, its successfully combined solution-processing with a highly stable gallium-rich IGZO TFT.

VII. CONCLUSION

It's been succeeded in fabricating a gallium-rich IGZO through solution processes andcompared it to an indium-rich IGZO when subject to positive stress bias. Through tests and results, itsobserved that the stabilities of the devices and visualize/quantify their differences. Also found that the gallium-rich IGZO possessed 25-40 times the stability of the indium-rich device. The gallium-rich device lacked mobility relative to the indium-rich device but surpassed it in all other performance metrics such as V_{TH}, ΔV_{ON}, on/off ratio, SS, andstability. The good electrical characteristics of this device demonstrate the viability of our solution-processing method in fabricating working gallium-rich 2:2:1 IGZO TFTs. With this success in solution-processing, this 2:2:1 IGZO can be fabricated on a much larger scale with simple and cheap processes, making it a valuable result for display technology. Due to its high gallium content, its stability was impressively good, showing less than 1 V of shift through 10000 seconds of stress, making it a successful high-stability device.

2:2:1 IGZO TFT's high stability and good performance makes it an applicablematerial to be used in commercialized display devices such as OLED, as they are expected to perform consistently over time with good mobility. Not only that, but its simple fabrication makes it attractive to industries that might want to reduce costs and simplify fabrication. These results establish the template of a high-stability solution-processed IGZO TFT whose performance metrics such as mobility and switching characteristics can be further improved with more research. The stability of the device should be encouraging and spur more work done on the rarely reported solution-processed gallium-rich IGZO. With future work, our gallium-rich IGZO could not only improve in stability but also in all other electrical characteristics and replace existing technologies as its full potential is explored.

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