

Comparison of composition and atomic structure of amorphous indium gallium zinc oxide thin film transistor before and after positive bias temperature stress by transmission electron microscopy

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Abstract

In this paper high resolution transmission electron microscopy analysis is performed on indium gallium zinc oxide thin film transistors to determine the crystal structure of the material. The relative elemental concentrations of indium, gallium, zinc and oxygen were quantified and analyzed using energy dispersive spectroscopy before and after subjection to positive gate bias temperature stress at 80 °C. Notable changes in the concentration of oxygen in the device channel were observed along with a reduced concentration of the elements indium, gallium and zinc after electrical stressing. We speculate this relative reduction in metal concentration could be attributed to the outdiffusion of metal ions from the channel region into the surrounding thermal oxide and the increase in the oxygen concentration in the stressed device is related to electric field assisted adsorption of oxygen from the ambient.

Keywords: thin films, IGZO, HRTEM, EDS

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

1. Introduction

In the next generation of flat panel displays and optical technologies, the ability to make electronic devices from transparent to near infrared and visible wavelengths is gaining importance. Amorphous indium gallium zinc oxide (a-IGZO) has been found to be a possible alternative to the already mature amorphous silicon thin film transistor technology used in display applications. IGZO thin film transistors (TFTs) are better than their predecessors in terms of carrier mobility. High resolution transmission electron microscopy (TEM) analysis of a-IGZO has provided evidence of local atomic clustering after annealing the amorphous structure [1]. Energy dispersive x-ray spectroscopy (EDS) of a-IGZO has revealed electro migration of the constituent elements, especially indium, resulting in an

increase in the threshold voltage of the fabricated TFT. Momentum transfer by the injected electrons was speculated to be the most possible mechanism [2]. The conductivity of the IGZO film is mainly governed by the 5s orbitals of In ions forming the conduction band minimum, while the overall performance can be attributed to the relative concentration of the component elements, which have different contributions to the electrical properties of the film [2]. Interstitial oxygen atoms, which occupy the octahedral interstitial sites, can influence the device performance by behaving as acceptors. EDS analysis of an as-deposited amorphous IGZO has confirmed that the region closest (10–14 nm) to the interface is richer in oxygen as compared to the remaining regions, which is attributed to oxygen diffusion [3]. In general, more oxygen deficiency was observed after thermal annealing [3]. Bias

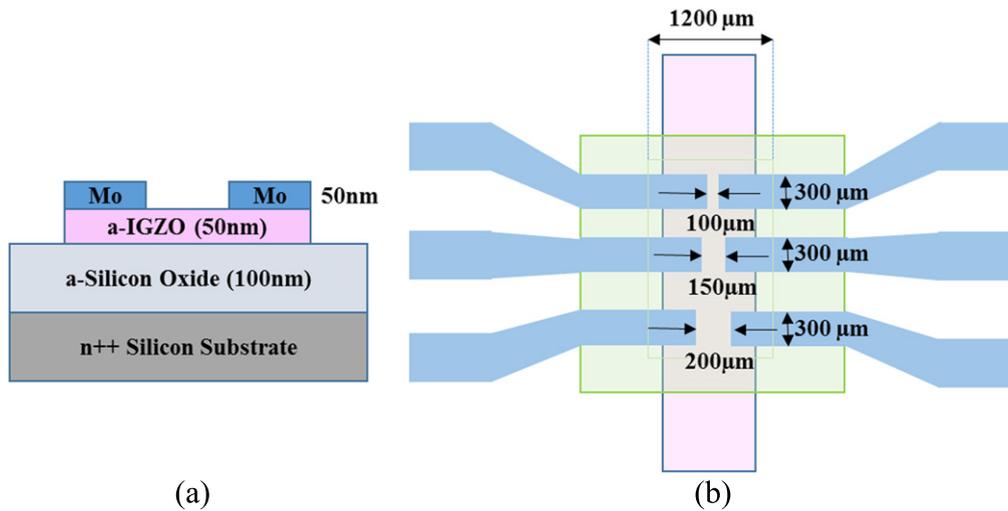


Figure 1. (a) Shows the thin film transistor cross section schematic and (b) shows the layout.

temperature stress (BTS) experiments conducted on a-IGZO TFT have shown a positive shift in the threshold voltage under positive bias and a negative shift on application of a negative bias. A 75% reduction of BTS degradation was reported with a post thermal annealing step. Although the exact nature of this reduction was unclear, the reduction in the oxygen content in the bulk and interfacial layer was speculated to be the main reason [4]. In this paper, we have performed high resolution transmission electron microscopy and EDS analysis of sputtered amorphous IGZO TFTs before and after positive BTS at 80 °C. The effect of the positive gate bias temperature stress on the concentrations of indium, gallium, zinc and oxygen are analyzed using the EDS quantification results.

2. Experimental details

2.1. Device fabrication

A thermal oxide of thickness 100 nm was grown over an n++ crystalline substrate, with the n++ substrate being used as the gate electrode. An active channel layer of IGZO of thickness 50 nm was sputtered onto the oxide using dc sputtering using an InGaZnO target with a composition of In:Ga:Zn:O in 2:2:1:7 atomic ratio. The dc sputtering was carried out at a power of 200 W, pressure of 4 mTorr and gas flows of 1.5 sccm O₂ and 30 sccm Ar. Molybdenum was chosen as the metal source/drain contact to the a-IGZO since it forms a stable ohmic contact with the film owing to a work function of around 4.36 eV. Molybdenum was sputtered at 600 W dc + 22 W RF power, pressure of 3.78 mTorr and an argon gas flow of 40 sccm using a Kurt J Lesker sputtering system. Shadow masks were used to define the source and drain contacts. The TFT was annealed at 350 °C for 30 min. The entire fabrication of the TFT was carried out in the Lurie Nanofabrication Facility (Department of Electrical and Computer Engineering) at the University of Michigan, Ann Arbor.

Thin film transistors were fabricated with channel lengths of $L = 100, 150$ and $200 \mu\text{m}$ and a channel width of $300 \mu\text{m}$.

One of the devices was electrically degraded on being subjected to positive gate bias temperature stress at 80 °C while the other was a virgin device. The channel area was devoid of metal electrodes. The analysis was done on the area at the center, i.e., $L = 150 \mu\text{m}$ for each device. Figure 1 shows the schematic cross section and the layout of the thin film transistor used in this study.

2.2. TFT electrical testing

One of the devices that was fabricated, as mentioned earlier, was subjected to positive gate bias temperature stress at 80 °C. To collect the TFT transfer characteristics, the gate of the thin film transistor was swept from -10 V dc to $+10 \text{ V dc}$. The measurements were taken at drain–source voltage V_{ds} namely 0.1 V dc . The TFT used in this experiment has a threshold voltage of around -5.8 V and a mobility of $30 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at a temperature of 80 °C.

The initial measurements were taken using a reference of 0 s. The TFT was stressed for a stress time ranging from 0, 100, 400, 700, 1000, 4000, 7000 to 10 000 s by applying a voltage of $+20 \text{ V}$ at the gate terminal. During the stress period, the temperature of the sample was raised to 80 °C. These measurements were carried out in the dark using Agilent semiconductor parameter analyzer. Figure 2 shows the TFT transfer characteristics at 80 °C in linear region of operation. As shown in the graph, the subsection of positive bias temperature stress resulted in the transfer characteristics shifting to the right. This means that the threshold voltage of the device is increasing. A possible speculation for this phenomenon is negative oxygen concentration increasing in the amorphous IGZO layer, which in turn results in the increase of the threshold voltage [5].

2.3. TEM sample preparation

The cross-sectional TEM samples were prepared using an FEI Quanta 200 3D Dual Beam focused ion beam (FIB) using the ‘*in situ* lift-out’ technique. This technique utilizes the equipment’s precision milling capability at the specified region of interest, which is the center between the source and drain

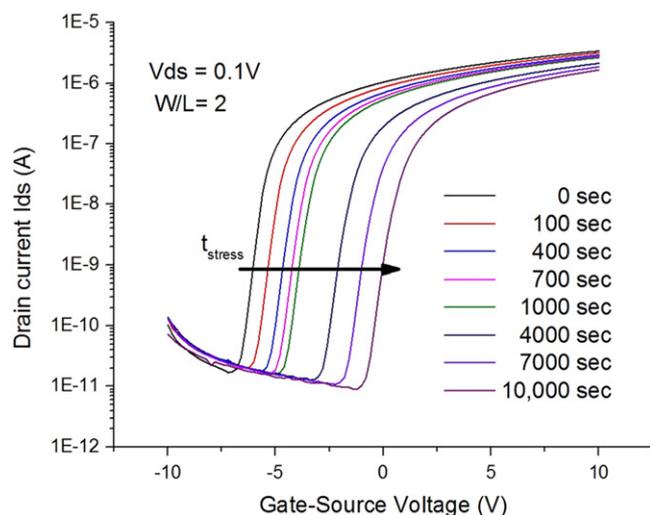


Figure 2. a-IGZO TFT transfer characteristics at 80 °C are shown.

electrodes for our samples with good accuracy (as shown in figure 3).

A layer of carbon was first deposited using the FIB over the TFT structure to prevent damage during the ion beam milling. The FEI Quanta 200 3D, which is a dual-beam system consists of a tungsten source electron beam column and a Gallium ion beam column.³ Both the electron and ion beams can be used to deposit materials using gas injectors that contain various precursor gases which decompose under the beam. The electron beam is also used for imaging and can be operated under a range of accelerating voltages and currents depending on the conductivity of the sample.

The ion beam is primarily used for milling and two large trenches are first milled on either side of the carbon that had been previously deposited using a large beam current and 30 kV accelerating voltage. A smaller beam current is then used to reduce the thickness of this membrane to approximately one micron thickness. The final milling voltage was decreased to 5 kV to further thin the samples and minimize sidewall damage incurred from the 30 kV ion beam.

The thin membrane is milled until the final thickness is approximately 100 nm. At these dimensions, the membrane is transparent to electrons and can hence be used for analysis. For lift-out, the sample is directly attached to a needle which is operated using a high precision micromanipulator and then attached onto the supporting TEM grid using FIB deposited platinum.

2.4. Analysis of device structure

2.4.1. Transmission electron microscopy. High resolution TEM and scanning transmission electron microscopy (STEM) were performed using an FEI Titan G2 TEM equipped with a spherical aberration corrector on the probe-forming lens at an accelerating voltage of 200 kV. The aberration corrector allows for sub-angstrom resolution to be achieved during

imaging. The electron source in the Titan is an X-FEG Schottky emitter which produces a highly coherent and bright electron beam with good emission stability. High resolution TEM relies on phase contrast of the scattered electrons and allows us to obtain images that are representative of the atomic structure of the material. TEM imaging uses a parallel electron beam as opposed to STEM mode, in which the beam is strongly converged down to a point on the sample and then rastered across a square area. When performing STEM imaging on these samples, the individual point dwell times were kept low, at 15 μ s, as electron beam induced indium diffusion into silicon oxide was observed for longer dwell times. STEM images were collected using a high angle annular dark field (HAADF) which results in an image that primarily shows mass contrast, approximately proportional to the square of the atomic number of the elements.

2.4.2. Energy dispersive x-ray spectroscopy. While operating the microscope in STEM mode, EDS was performed simultaneously in order to compare the relative composition of the two samples. A sample exposed to a focused beam of electrons generates x-rays which have a unique spectrum that is characteristic of the elements present in the sample. Quantitative analysis of the spectra includes measuring the line intensities for each element in the sample. This is done by first performing a background calculation and subtraction, then a peak deconvolution process, and the final quantification. EDS maps were acquired in the Titan G2 TEM using ChemiSTEM quad detectors at a beam current of 0.6 nA. ChemiSTEM technology uses the combination of the X-FEG source and Super-X x-ray detectors which are based on silicon drift detector technology to perform ultra-fast EDX mapping. These detectors have a total area of 120 mm² and a solid angle of collection of 0.9 sr. Due to its high collection area, the Super-X integrated EDX system provides very sensitive detection of most elements including oxygen since it is a windowless detector [6]. Standard less Cliff–Lorimer quantification was done on the deconvoluted EDS line intensity data using the Bruker Espirit software. Deconvolution is necessary because it eliminates the overlap of different intensities and peaks causing misidentification and problems with quantification. Although the absolute accuracy of EDS composition determined by standard less quantification methods can be very large, the relative accuracy when comparing samples with data collected under identical conditions is much smaller and should be within a few percent. The spectrum obtained by EDS analysis is quantified by the Cliff–Lorimer ratio method to obtain the relative concentrations between the elements from the EDS peak intensities [7].

3. Results and discussion

3.1. High resolution electron microscopy results

Figure 4 shows the HRTEM images obtained after performing the analysis using the Titan G2 instrument. Figure 4(b) shows

³ The data is acquired from the open source of the company FEI (www.fei.com)

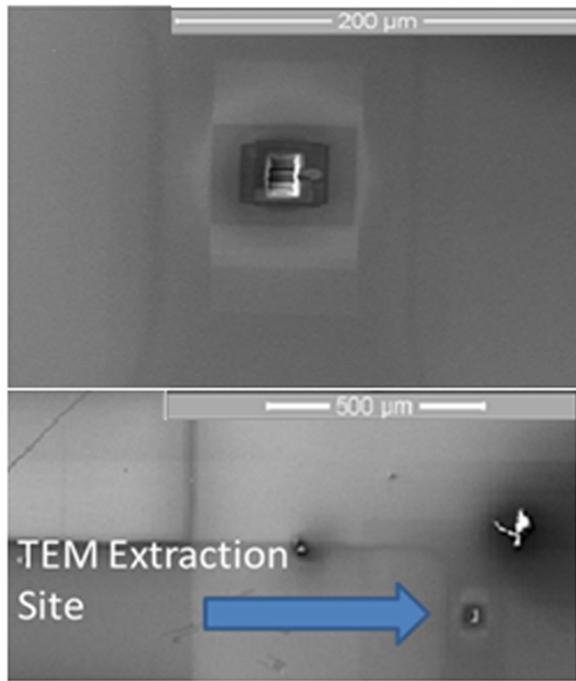


Figure 3. TEM extraction site on the sample is shown including a magnified image.

reference crystalline silicon substrate with grown thermal oxide on the top. We can see the interface between the silicon substrate and the thermal oxide grown over it at a very high resolution which allows us to easily see the lattice fringes in the crystalline silicon substrate. The thermal oxide, however, does not show any lattice fringes in the HRTEM image, thereby confirming that the material is amorphous. The interface between silicon and silicon oxide appears to be abrupt with no interfacial layer between the two layers. Figure 4(a) shows the interface between not electrically stressed amorphous indium gallium zinc oxide and the thermal oxide collected under the same experimental conditions. The a-IGZO layer does not show the presence of any crystal orientation like the silicon substrate at the same resolution. Hence, the InGaZnO sputter deposited layer is amorphous before and after BTS under the used experimental conditions. Also, the interface between a-InGaZnO and thermal oxide does not appear to be as abrupt when compared to the interface between thermal oxide and silicon. However this may be due to indium diffusion into the silicon oxide under longer dwell times as was observed, or because of the possibility of a slight tilt of the electron beam to the interface. The atomic force microscopy surface roughness measurements on the silicon oxide confirmed that the surface was smooth with average roughness of $R_a = 0.251$ nm and root mean square surface roughness of $R_{ms} = 0.307$ nm, and thus the interface linescan broadening cannot be attributed to the surface roughness of thermal silicon oxide surface.

Figure 4(c) shows the Fourier transform (FFT) of the HRTEM image of InGaZnO thin film. Regular periodicities, i.e., lattice fringes, in the image would produce a spot in reciprocal space. The diffuse pattern of the FFT and lack of

discrete spots indicates that there is no observed crystallinity in the sputter deposited IGZO. We can hence conclude that our sputter deposited IGZO film is indeed amorphous. Also, the circular nature of the FFT taken from the HRTEM image confirms that the alignment of the TEM was good in this experiment.

3.2. Energy dispersive x-ray spectroscopy results

The EDS analysis performed on the FEI Titan G2 TEM provided some interesting results. It was performed on both the virgin device and the device subjected to positive gate bias temperature stress at 80 °C. The quantification done by the software produced the following results tabulated below.

Table 1 shows the experimental concentrations in normalized weight and normalized atomic percent of oxygen, gallium, zinc and indium in the virgin device. The summation of the percentage concentrations in the sample is 100. Table 2 shows the concentrations in normalized weight percentage and normalized atomic percentage of oxygen, gallium, zinc and indium in the device that was subjected to positive bias temperature stress at 80 °C. Normalization is done by dividing the concentration of a particular element by the summation of the concentrations of all the different constituent elements. We can compare the normalized atomic percentage of the concentrations between the virgin device and the electrically degraded device to conclude that the relative concentration of oxygen in the degraded device has significantly increased in comparison to the virgin device relative to the other elements gallium, indium and zinc. The concentration of zinc is decreased by around 2.8%, but with an error in measurement of 1% this value could not be very significant. The normalized atomic percentage concentration of indium is relatively constant in both cases. The normalized atomic percentage of gallium has decreased by 4.5% from the virgin to the electrically degraded device. However, the error in measurement is approximately 2.7%. Hence, this reduction is not as significant when compared to oxygen increase. Since there is a reduction in the metal ion concentration in the stressed device in comparison with the virgin device, a possible explanation would be of the diffusion of the elements indium, gallium and zinc into the adjacent thermal oxide on stressing the device and the increase in the oxygen concentration could be attributed to the electric field assisted adsorption of the oxygen from the ambient to form oxygen species like O^{2-} or O^- . The threshold voltage shift on subjection to positive gate bias temperature stress at 80 °C can be accredited to the formation of these oxygen species [5].

As we have concluded that oxygen is the primary element whose relative concentration changes rapidly in amorphous IGZO, we shall look at the EDS quantification maps that were produced for the element oxygen at a magnification of 1.3 million in figure 5 which shows the EDS maps for the virgin device and the device subjected to positive bias temperature stress at 80 °C. These x-ray maps provide information about elemental distributions in the sample. Elemental x-ray EDS maps provide a representation of the number of x-rays detected for a given element by converting that into a

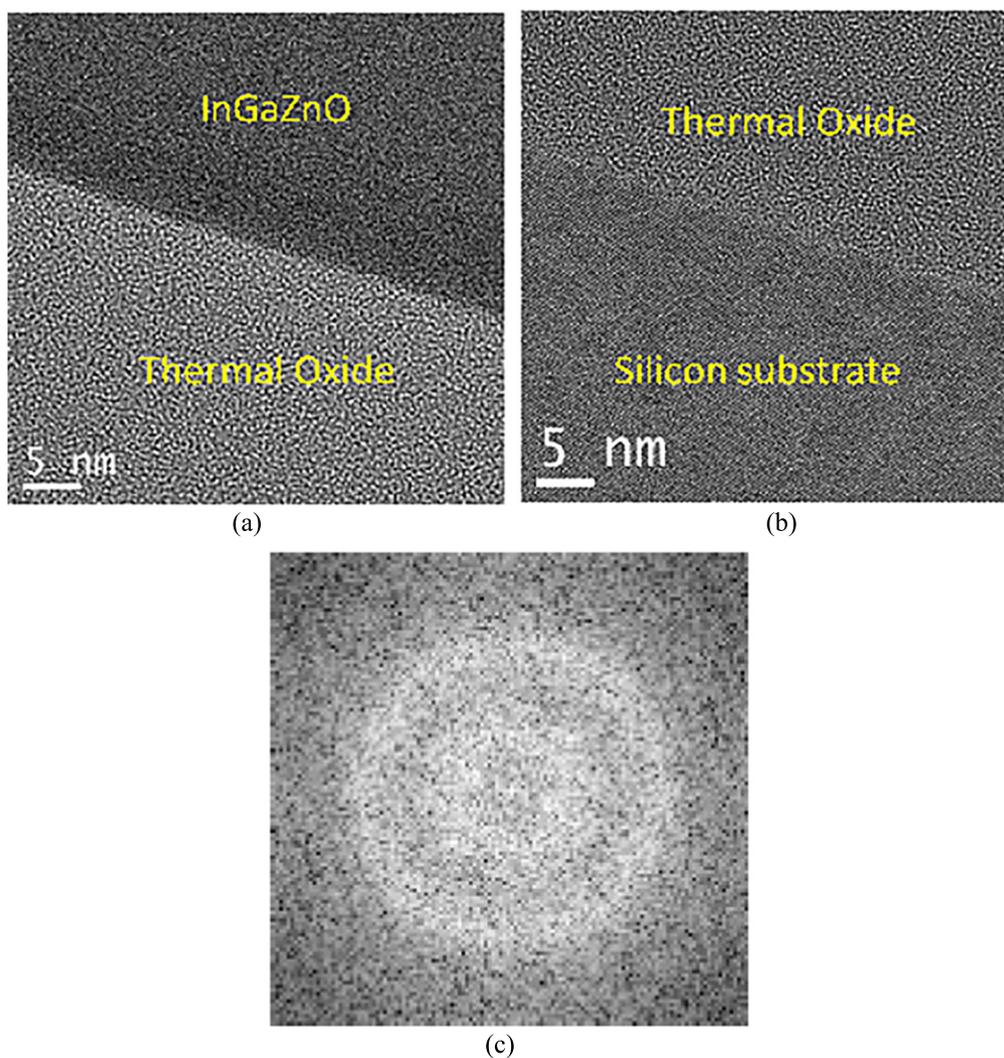


Figure 4. HRTEM images of (a) a-InGaZnO/thermal silicon oxide and (b) of thermal silicon oxide/Si interface are shown along with the FFT of the HRTEM image of IGZO in (c).

Table 1. EDS quantification results for the virgin device are shown.

Element	[Norm.wt.%]	[Norm at.%]	Error in t.%
Oxygen	11.7	41.8	1.1
Gallium	30.5	24.9	2.9
Zinc	12.5	10.9	1.2
Indium	45.3	22.4	13.7
Sum	100	100	

Table 2. EDS quantification results for the electrically degraded device are shown.

Element	[Norm.wt.%]	[Norm at.%]	Error in t.%
Oxygen	14.8	49.1	1.4
Gallium	26.7	20.4	2.5
Zinc	9.9	8.1	1.0
Indium	48.6	22.5	14.7
Sum	100	100	

brightness value for each pixel in the digital image. These maps show the distribution of oxygen in red across the a-IGZO/SiO₂ interface at a high resolution.

Figure 6 shows the position where the linescans were extracted from the virgin and the electrically degraded thin film transistor, respectively. Figure 7 shows the linescan obtained from EDS maps of atomic percent for the elements across the layer versus position for the virgin device. The interfaces between the FIB protective layer, amorphous IGZO and the thermal oxide are approximately shown in the figure. However, since both the silicon oxide and the IGZO layer are amorphous, we cannot be sure if the beam is exactly parallel to the interface. A small tilt could result in the spreading of the apparent compositional profile at the interfaces. Lastly, there is the uncertainty in the degree of electron beam induced diffusion across the interface. Although the dwell time was minimized in order to prevent any obvious beam induced diffusion, it is still possible that some may have occurred at the interface. Because of these reasons, it is hard to say with

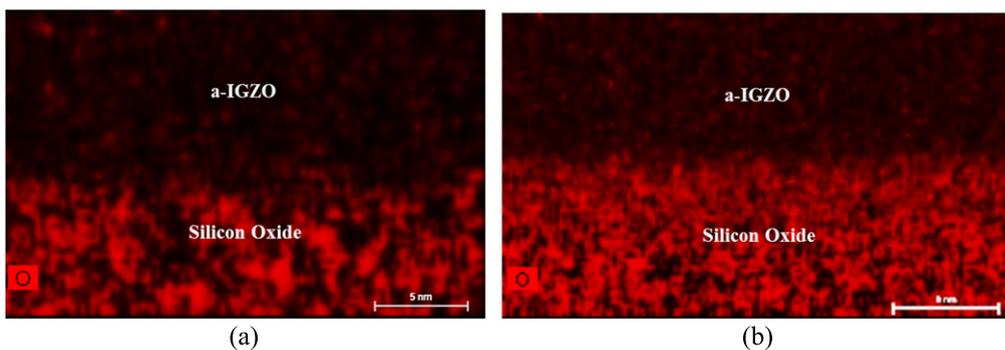


Figure 5. EDS Maps for a-IGZO/SiO₂ interface for (a) the virgin device and (b) the electrically degraded device at atomic resolutions of 5 and 8 nm, respectively.

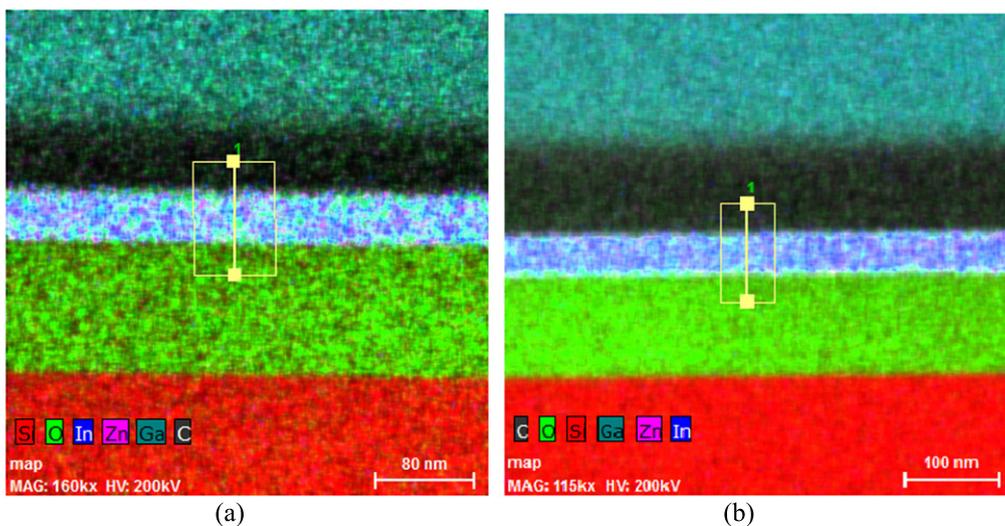


Figure 6. Linescan locations for (a) virgin device and (b) electrically degraded sample are shown.

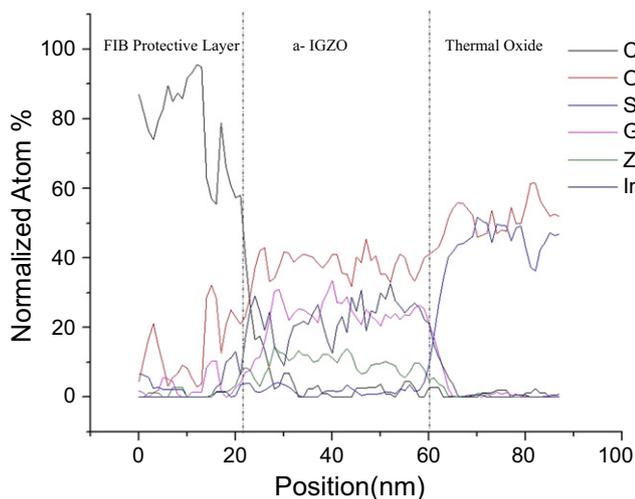


Figure 7. EDS linescan of the virgin device.

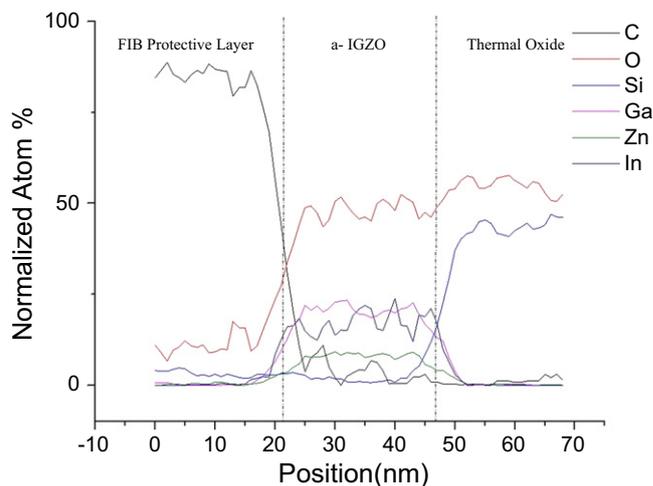


Figure 8. EDS linescan of the electrically degraded device.

absolute certainty about the degree of diffusion of the metal ions at the interface.

Figure 8 shows the linescan obtained from the EDS maps for the device subjected to positive bias temperature stress at 80 °C. The interfaces between the FIB protective layer, amorphous IGZO and the thermal oxide are approximately

shown in the figure. Thus, comparing figure 7 with figure 8, we can see that the relative concentration of oxygen has increased significantly and there is a probability of diffusion of the elements indium, gallium and zinc out of the InGaZnO layer on the subjection of a positive bias temperature stress. Since both samples were subjected to identical TEM electron

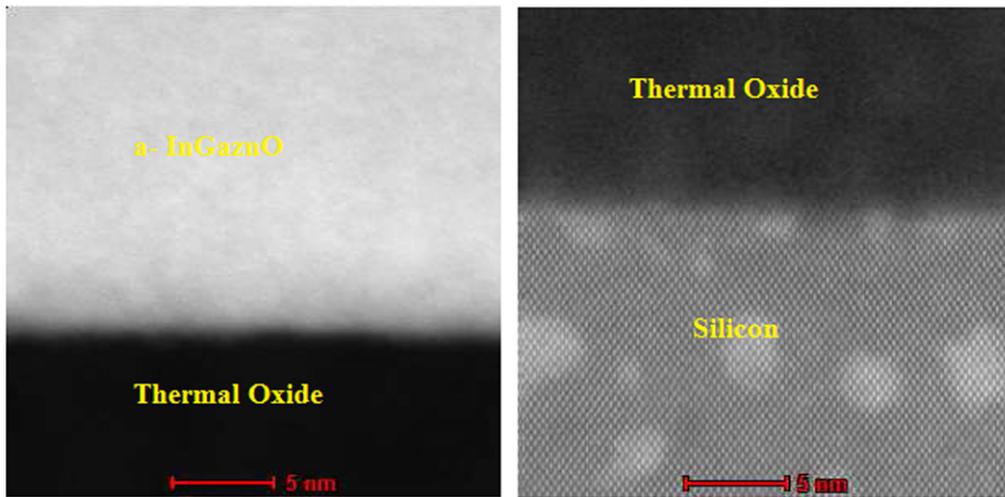


Figure 9. HAADF images of the thin film virgin device are shown.

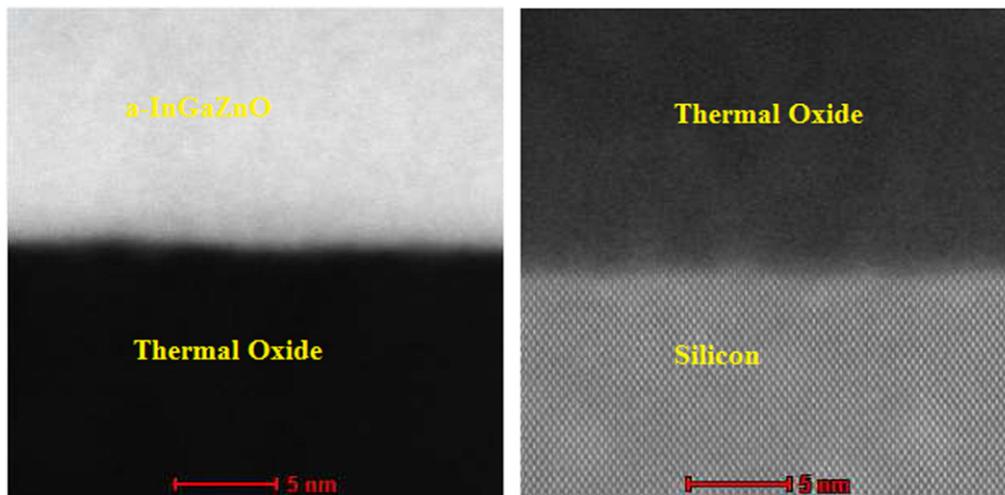


Figure 10. HAADF images of the electrically degraded thin film device are shown.

beam exposure times, we would expect any beam induced diffusion to be similar for both samples. Therefore, the significant difference in relative oxygen concentration is likely due to actual differences between the samples.

3.3. Scanning transmission electron microscopy results

STEM images of the thin film transistor support our analysis from the HRTEM results. High angle annular dark field images obtained in STEM mode show a large difference in contrast between the IGZO and Si oxide due to the composition difference. STEM images from the silicon substrate interface demonstrate that atomic resolution is obtained under these imaging conditions in STEM mode. Some FIB re-deposition is observed because of the sample preparation method employed. From figure 9, we can see that the interface between the amorphous IGZO and the thermal oxide does not appear to be abrupt. Figure 10 shows the HAADF images obtained for the thin film transistor subjected to positive gate bias (+20 V) temperature stress at 80 °C which appear to be similar.

4. Conclusion

We have performed high resolution transmission electron spectroscopy and energy dispersive spectroscopy on amorphous IGZO TFT device structures. We found that the sputter deposited layer is amorphous from the high resolution TEM and corresponding FFT images under the used experimental conditions. The scanning transmission electron microscopy images indicate that the interface between amorphous InGaZnO and thermal oxide is not as sharp and abrupt as compared to the interface between crystalline silicon substrate and thermal oxide. The energy dispersive spectroscopy analysis of the amorphous IGZO layer indicated a relative oxygen content increase in the stressed device. This increase is attributed to the electric field adsorption of oxygen species such as O^{2-} or O^- on the application of a positive gate bias temperature stress at 80 °C. Outdiffusion of the elements indium, gallium and zinc is also speculated which is correlated to the decrease in their respective concentrations after being subjected to positive bias temperature stress.

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