



Article Effect of Hydrogen Migration in SiO₂/Al₂O₃ Stacked Gate Insulator of InGaZnO Thin-Film Transistors

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Abstract: In this work, the correlation between SiO_2 deposition thickness and hydrogen content is discussed and the effect of the SiO_2 layer on the properties of synaptic InGaZnO (IGZO) TFTs is analyzed. Three types of IGZO synaptic thin-film transistors (TFTs) were fabricated with different gate insulators, and the effect of SiO_2 as a gate insulator was investigated. XPS analysis confirmed that the hydrogen content in the Al_2O_3 and SiO_2 layers increased during SiO_2 deposition step for all depth regions. Hydrogen injected by the SiO_2 layer deposition step was confirmed to improve the memory window through more threshold voltage shift under positive bias stress (PBS) and negative bias stress (NBS) conditions. In addition, the retention characteristics were improved due to the low hydrogen movement velocity in the SiO_2 layer. These results contribute to the optimization of the amount of hydrogen, and the proposed device has potential as a synaptic device capable of neuromorphic computing.

Keywords: synaptic device; InGaZnO thin-film transistor; hydrogen; low-temperature atomic layer deposition; XPS

1. Introduction

It is necessary to efficiently process a large amount of data as advanced technologies related with artificial intelligence such as autonomous vehicles have recently approached to our lives. In the case of conventional von Neumann computing systems including central processing units, a bottleneck occurs when processing a large amount of data due to a serial computation and a physically separated structure between the memory and the processor [1]. Neuromorphic systems are enabled to perform vector-by-matrix-multiplication (VMM) operations in parallel using Ohm's law and Kirchhoff's law, which enables low-power and high-efficient in-memory computing [2–8]. Therefore, it is important to investigate synaptic devices suitable for neuromorphic systems in artificial neuromorphic computing systems.

Recently, in addition to two-terminal synaptic devices such as resistive switching memory [9–16] and phase change memory [17–20], three-terminal synaptic devices are also receiving great attention because input and output ports can be used independently [21–29]. Among them, InGaZnO (IGZO) thin-film transistors (TFTs) has good compatibility with conventional processes and can be utilized as a nonvolatile memory. It is also widely used especially in flexible applications and displays thanks to its low-temperature processing, uniformity, low-leakage current and high mobility [30–34]. Therefore, it is necessary to verify the synaptic properties by integrating IGZO TFTs on a flexible substrate using a low-temperature process.

In this study, we fabricated three types of synaptic IGZO TFTs (single Al_2O_3 layer and double stack layers Al_2O_3/SiO_2 with SiO_2 thickness of 5 nm and 10 nm, respectively) on flexible substrates using different stacks of gate insulators. The devices threshold voltage (V_T) modulation can be achieved under positive gate bias stress (PBS) and negative gate



Citation: Park, S.; Youn, S.; Jang, J.T.; Kim, H.; Kim, D.H. Effect of Hydrogen Migration in SiO₂/Al₂O₃ Stacked Gate Insulator of InGaZnO Thin-Film Transistors. *Crystals* **2022**, *12*, 594. https://doi.org/10.3390/ cryst12050594

Academic Editors: Francisco M. Morales and Simona Binetti

Received: 29 March 2022 Accepted: 20 April 2022 Published: 23 April 2022

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Copyright: © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). bias stress (NBS) due to the movement of hydrogen ions in the Al_2O_3 layer deposited by low-temperature (LT) atomic layer deposition (ALD). In addition, X-ray photoelectron spectroscopy (XPS) analysis and electrical measurement demonstrated that the SiO₂ layer deposited by e-beam evaporation was a source of additional hydrogen ions and improved the memory window. Lastly, it was confirmed that the SiO₂ layer contributed to the improvement of retention by controlling the movement velocity of hydrogen ions.

2. Materials and Methods

Three types of IGZO TFTs were fabricated on a polyethylene terephthalate (PET)/SiO₂/p⁺-Si substrate with varying the gate dielectric stack as show in Figure 1a–c. Except the gate insulator deposition, A-type (Al₂O₃ layer), B-type (Al₂O₃/SiO₂ 5 nm) and C-type (Al₂O₃/SiO₂ 10 nm) devices share the whole process flow. First, A PET film was cleaned with acetone in a sonicator. On the PET substrate, 50-nm-thick SiO_2 layer was deposited using an e-beam evaporator as a buffer layer. The gate layer was deposited with 20 nmthick Cu and patterned by e-beam evaporation and lift-off, respectively. The single layer (without SiO₂) of Al₂O₃ (40 nm) and double layer of Al₂O₃ (40 nm)/SiO₂ (5 and 10 nm) were subsequently deposited by LT ALD (Al_2O_3 layer) with Al(CH₃)₃ and H₂O as reactant vapor at 80°C and e-beam evaporator (SiO₂ layer), respectively. Then, 35-nm-thick IGZO layer was deposited at a room temperature with an active layer shadow mask by reactive sputtering $(3(Ar)/0.1(O_2)$ sccm, 5 mTorr, 150 W RT power). The source and drain regions were formed with 40-nm-thick Cu deposited by an e-beam evaporator followed by lift-off. Finally, the devices were annealed in air for 1 h at 250 °C in the air condition for detaching PET substrate from the silicon substrate. The channel width and length of the devices were defined as 50 and 20 µm, respectively.



Figure 1. Schematic view of three-type bottom gate IGZO TFTs. (**a**) LT ALD-deposited 40 nm Al₂O₃ (A-type), (**b**) LT ALD 40 nm Al₂O₃/e-beam 5 nm SiO₂ (B-type) and (**c**) 10 nm SiO₂ (C-type).

3. Results

Figure 2a–c show the I-V characteristics of three types of IGZO TFTs under PBS condition at gate voltage (V_{GS}) = 7 V. As the PBS time increases, the three types of IGZO TFTs show a negative V_T shift of 0.4, 0.6, and 2.2 V, respectively. The thicker the SiO₂ layer is, the larger V_T shift is obtained. In our previous study, the V_T modulation of IGZO TFT was determined by the migration of hydrogen ions inside Al₂O₃ layer [35,36]. In addition, when additional SiO₂ layer was deposited as a gate insulator on the Al₂O₃ layer, a larger V_T shift was obtained under PBS because additional SiO₂ deposition step helps to generate additional hydrogen by dissociating the Al-H bonds in Al₂O₃ layer. In our previous study, it was confirmed that the hydrogen content was much higher when the SiO₂ layer was deposited on the Al₂O₃ than when deposited without the Al₂O₃ layer [37]. This implies that the insertion of an additional SiO₂ layer acts as a hydrogen source, and the amount of hydrogen depends on the thickness of the SiO₂ layer. Additional hydrogen ions moving toward the IGZO channel under the PBS help to form an inversion layer of the IGZO channel and increase the amount of negative V_T shift since hydrogen acts as an n-type dopant [38,39].



Figure 2. I-V characteristics of (**a**) A-type, (**b**) B-type, and (**c**) C-type IGZO TFTs under a positive bias stress (PBS). When the thickness of the SiO₂ layer is sufficiently wide, a significant threshold voltage shift occurs in the PBS condition.

Figure 3a show the schematic view of hydrogen generation during the deposition of the SiO₂ layer. When the LT ALD process is carried out for the Al_2O_3 deposition, residual hydrogen H is generated inside the Al₂O₃ layer due to incomplete chemical reaction between Al(CH₃)₃ and H₂O [40,41]. Next, while the SiO₂ layer is deposited on the Al₂O₃ layer, additional hydrogen is injected from the Al_2O_3 to the SiO₂ layer. Figure 3b,c shows the XPS analysis of O 1 s in the SiO₂ layer for the B- and C- type devices, respectively. The amount of hydrogen is verified through the O1 s XPS spectrum, which is deconvoluted to three peaks of O-H, Vo, and M-O to analyze hydroxide, oxygen vacancy, and metal-oxygen content, respectively [42,43]. Hydrogen content in the SiO₂ layer can be confirmed in the form of hydroxide, and it can be also observed that the C-type device with a thicker SiO_2 has 20% more hydrogen than the B-type device. This is because a thicker SiO₂ layer means a longer deposition time, and the existing Al-H bonds are more dissociated and injected into the SiO₂ layer. The change of hydrogen content according to the SiO₂ thickness shows a clear difference in the Al₂O₃ layer as shown in Figure 3d–f. The difference in hydrogen content between A-type and B-type is only 6%, while the difference between the A-type and C-type is 31%. This means that the B-type with 5-nm-thick SiO₂ layer cannot generate significant hydrogen content compared to the C-type with SiO_2 10 nm layer, and the C-type can be more suitable for memory device considering the V_T modulation.

Moreover, we performed the additional XPS depth profile analysis of the entire stack for the IGZO TFTs to analyze the movement of hydrogen in detail during the gate insulator deposition step. Figure 4 summarizes the proportion of hydroxide in the total XPS depth profile analysis for the A-, B- and C-type devices. First, the hydrogen content in the IGZO channel of all the devices is lower than that of either the SiO_2 or Al_2O_3 layer for the entire depth region. This is because the bond dissociation energy of Si-H (318 kJ/mol) and Al-H (285 kJ/mol) is higher than In-H (243 kJ/mol), Ga-H (274 kJ/mol), and Zn-H (85 kJ/mol), so the additionally generated hydrogen in the Al₂O₃ layer during the deposition process is hard to move toward the IGZO channel [44,45]. In addition, since hydrogen combines with oxygen to form an O-H bond when hydrogen reaches the IGZO interface, a large amount of hydrogen cannot move into the IGZO channels. Inside the SiO₂ layer, the hydrogen content of the C-type device is much higher than that of the B-type device in all the regions. This is because the hydrogen content increases with the increased SiO_2 deposition thickness because the existing Al-H bond is broken and injected into the SiO_2 layer during the deposition step as discussed in Figure 3b,c. At 5 nm depth of the Al_2O_3 layer, the hydrogen content of the C-type is much higher than that of the A- and B-type devices. However, as the depth increases, there is no significant difference in the hydrogen content depending on the device type. This is because additionally generated hydrogen during the SiO₂ deposition step moved into the SiO₂ rather than the Al₂O₃ layer due to the difference in bond dissociation energy between Si-H and Al-H.

The stress-induced instability in the A- and C-type IGZO TFTs is also investigated by applying NBS and PBS at $V_{GS} = -15$ V, and 10 V, respectively, for 1.8 ks as shown in

Figure 5a,b. The C-type device is considered as potential candidate for synaptic device thanks to high hydrogen content and the A-type device is verified as reference device. The A-type IGZO TFT shows 3.6 V negative V_T shift under PBS and 2 V positive V_T shift under NBS, which means that hydrogen ions inside the Al₂O₃ and SiO₂ layer move toward the IGZO channel during PBS but move away from the IGZO channel again during NBS due to negative V_{GS} . Since the C-type IGZO TFT has more hydrogen content than the A-type device due to the additional SiO₂ layer, it shows a larger V_T shift under the same stress conditions ($\Delta V_T = -7.9 \text{ V/6 V}$ under PBS/NBS). The V_T is recovered through NBS in both the A- and C-type devices, but not fully as the initial values since some of hydrogen moving toward the IGZO layer can form a bond with oxygen at the IGZO layer interface [40], and some hydrogen can act as a shallow donor [46]. Therefore, strong NBS condition with larger amplitude than PBS condition is required for sufficient V_T recovery.



Figure 3. Illustration of hydrogen movement in the Al_2O_3 , SiO_2 layer during the gate insulator deposition. (a) Schematic view of the IGZO synaptic TFTs with hydrogen movement by deposition process. O 1 s XPS spectra of Al_2O_3 layer in (b) B-type and (c) C-type IGZO synaptic TFTs in the SiO₂ layer. O 1 s XPS spectra of Al_2O_3 layer in (d) A-type, (e) B-type, and (f) C-type IGZO synaptic TFTs in Al_2O_3 layer.



Figure 4. XPS depth profile analysis of A-, B- and C-type IGZO TFTs. XPS analysis was performed at 5 nm, 15 nm, 25 nm, 35 nm depth in IGZO layer (all the types), 5 nm, 10 nm depth in SiO₂ layer (B- and C-type only), and 5 nm, 10 nm, 25 nm, 40 nm depth in Al₂O₃ layer (all the types).



Figure 5. I-V characteristics of with PBS ($V_{GS} = 10 \text{ V}$, 1.8 ks) and NBS ($V_{GS} = -15 \text{ V}$, 1.8 ks) for (a) A-type and (b) C-type.

Figure 6a,b shows the retention characteristics after applying 16 potentiation pulses of $V_{GS} = 8$ V with a width of 100 ms to the A- and C-type IGZO TFTs, respectively. After all the potentiation pulses are applied, both the drain current of both devices is slowly recovered to the initial state, indicating that hydrogen move inside the Al₂O₃ and SiO₂ layer. However, the drain current of the A-type decreases rapidly, while that of the C-type remains higher than the initial value for 4 s after applying 16 potentiation pulses. The drain current of the A-type device decreased to almost the initial state (96% drop) for 4 s, but the C-type device maintained the drain current (27% drop) for 4 s. This is because the migration velocity of hydrogen in SiO₂ is slower than that in Al₂O₃ [47], which implies that the insertion of the SiO₂ layer improves not only the V_T modulation window but also the retention characteristics.



Figure 6. Retention characteristics of (**a**) A-type and (**b**) C-type devices after 16 potentiation pulses with $V_{GS} = 8$ V and 100 ms width.

4. Conclusions

In this study, we reported the effects of additional deposition of SiO_2 on the Al_2O_3 layer as a gate insulator of the IGZO TFTs. A total of three types of the devices were fabricated with different gate insulator stacks and their electrical characteristics were compared to investigate the effect of the additional SiO_2 layer. It was confirmed through the XPS analysis that hydrogen ions were additionally generated by inserting the SiO_2 layer between the Al_2O_3 and IGZO layers. With increasing the SiO_2 layer thickness, the V_T memory window was improved under PBS and NBS conditions thanks to the increased hydrogen content. In addition, the retention properties were significantly improved as the migration velocity of hydrogen in SiO_2 decreased. These results are expected to contribute to the optimization of the synaptic IGZO TFTs with LT processed gate insulators.

Author Contributions: Methodology, S.P. and J.T.J.; formal analysis, S.P. and S.Y.; writing—original draft preparation, S.Y.; writing—review and editing, H.K. and D.H.K.; project administration, H.K. and D.H.K.; funding acquisition, H.K. and D.H.K. S.P. and S.Y. equally contributed to this work. All authors have read and agreed to the published version of the manuscript.

Funding: This work was supported in part by the National Research Foundation of Korea (NRF) grant funded by the Korea Government (MSIT) (No. 2016R1A5A1012966, 25%, No. 2020R1A2B5B01001979, 25%, and No. 2020M3F3A2A01081656, 35%), partly by the Institute of Information and Communications Technology Planning and Evaluation (IITP) grant funded by the Korea government (MSIT) (No. 2021-0-01764, 15%), and in part by the Brain Korea 21 Four Program.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: The data presented in this study are available on request from the corresponding author.

Conflicts of Interest: The authors declare no conflict of interest.

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