Marked improvement in reliability of 150 °C processed IGZO thin-film transistors by applying hydrogenated IGZO as a channel material

Daichi Koretomo1, Shuhei Hamada2, Marin Morì2, Yusaku Magari1, and Mamoru Furuta1,2,3

1Engineering Course, Kōchi University of Technology, Kōmi, Kōchi 782-8502, Japan
2Material Science and Engineering Course, Kōchi University of Technology, Kōmi, Kōchi 782-8502, Japan
3Center for Nanotechnology, Research Institute, Kōchi University of Technology, Kōmi, Kōchi 782-8502, Japan

E-mail: furuta.mamoru@kochi-tech.ac.jp

Received April 21, 2020; revised May 8, 2020; accepted May 19, 2020; published online June 5, 2020

A marked improvement in the reliability of a high-mobility In–Ga–Zn–O (IGZO) thin-film transistor (TFT) is presented after 150 °C annealing by applying hydrogenated IGZO (IGZO:H) as a channel. To enhance field-effect mobility ($\mu_{FE}$), the atomic ratio of In:Ga:Zn was chosen to be 6:2:1. The IGZO:H TFT exhibited a $\mu_{FE}$ of 18.9 cm² V⁻¹ s⁻¹ without hysteresis. Moreover, the reliability of the IGZO:H TFT significantly improved after 150 °C annealing as compared with that of a conventional IGZO TFT. Thus, the use of IGZO:H is an effective method of improving both the electrical properties and reliability of TFTs for flexible electronics. © 2020 The Japan Society of Applied Physics

Oxide semiconductors (OSs) represented by In–Ga–Zn–O (IGZO) are promising candidates for the active channel of flexible thin-film transistors (TFTs) since the field-effect mobility ($\mu_{FE}$) of OS TFTs is more than 10 times as high as that of amorphous Si TFTs. However, OS TFTs generally require thermal annealing at ~300 °C to reduce defects formed as a result of various types of processing damage. Several approaches such as simultaneous UV and thermal treatments, wet-O₂ annealing, and high-pressure gas annealing have been proposed to reduce the annealing temperature for flexible devices. Our group reported that defects generated in as-deposited IGZO films could be reduced through the low-temperature (150 °C) annealing of hydrogenated IGZO (IGZO:H) deposited by sputtering in an Ar + O₂ + H₂ gas mixture. A $\mu_{FE}$ of 13.4 cm² V⁻¹ s⁻¹ was achieved for a TFT with an IGZO:H (In:Ga:Zn = 1:1:1 at%) channel after 150 °C annealing. An improvement in the $\mu_{FE}$ of OS TFTs has been required to widen their range of application. However, the reliability of a high-mobility OS TFT significantly deteriorates when the annealing temperature is reduced from 350 to 150 °C. Thus, performance and reliability improvements are still challenging issues for TFTs with low thermal budgets.

In this letter, the effects of oxygen and hydrogen ratios during IGZO:H deposition are discussed as a means of enhancing the performance and reliability of TFTs through low-temperature annealing at 150 °C. It was found that the reliability of an IGZO:H TFT significantly improves after 150 °C annealing in comparison with that of an IGZO TFT with a conventional Ar + O₂-sputtered IGZO channel.

Bottom-gate IGZO TFTs were fabricated on a heavily doped n⁻-Si substrate with a 100-nm-thick thermally grown SiO₂ layer. The Si substrate and SiO₂ layer were used as the gate electrode and gate insulator (GI) for the TFTs, respectively. A 20-nm-thick IGZO channel was deposited by RF magnetron sputtering without substrate heating using a ceramic target of IGZO (In:Ga:Zn = 6:2:1 at%). IGZO621 was deposited with an Ar + O₂ gas mixture, while hydrogenated IGZO621 (denoted hereafter as IGZO621:H) was deposited with an Ar + O₂ + H₂ gas mixture. The O₂ and H₂ gas flow ratios, defined as $R$(O₂) = O₂/(Ar + O₂ + H₂) and $R$(H₂) = H₂/(Ar + O₂ + H₂), respectively, were varied under a total gas flow of 10 sccm. After the formation of an IGZO channel using a shadow mask, the IGZO channel was annealed at 150 °C in ambient air for 1 h. Then, a stacked film of Mo/Al/Mo was deposited by RF magnetron sputtering as source/drain electrodes using a shadow mask. Finally, post-annealing was carried out at 150 °C in ambient air for 1 h. The maximum processing temperature for the TFTs was 150 °C.

First, the annealing temperature ($T_a$) dependence of the Hall carrier density ($n_e$) of both the 30-nm-thick IGZO621 and IGZO621:H films was measured by changing the $R$(O₂) and $R$(H₂) values.

Figure 1(a) shows the $n_e$ of the IGZO621 films deposited at various $R$(O₂) values without H₂ gas as a function of $T_a$. For the as-deposited IGZO621 films, the $n_e$ of 2.0 × 10²⁰ cm⁻³ decreased to a value below the measurement limit (1.0 × 10¹⁶ cm⁻³) upon increasing $R$(O₂) to 10% or higher. However, the $n_e$ of the film deposited at $R$(O₂) of 10% increased by over three orders of magnitude (~10¹⁹ cm⁻³) with annealing at a $T_a$ of 150 °C. This result indicates that the IGZO621 film is thermally unstable because oxygen vacancies are easily formed in high-In-content OS films upon annealing. Moreover, an $R$(O₂) of more than 10% is required to obtain an appropriate $n_e$ for TFT fabrication with annealing at 150 °C.

Figure 1(b) shows the $n_e$ of the IGZO621:H films deposited with various $R$(H₂) values and a constant $R$(O₂) of 10%. The $n_e$ of the as-deposited IGZO:H film increased to more than 10¹⁹ cm⁻³ for the films deposited with $R$(H₂) values of 5% and 9% because hydrogen in IGZO acts as a shallow donor. On the other hand, the $n_e$ of the films deposited with $R$(H₂) values of 5% and 9% decreased by approximately two orders of magnitude (~3 × 10¹⁷ cm⁻³) after annealing at a $T_a$ of 150 °C. The $n_e$ of the films remained almost the same up to a $T_a$ of 350 °C.

Figure 1(c) shows the $R$(O₂) dependence of the $n_e$ of the IGZO621 and IGZO621:H films after annealing at a $T_a$ of 150 °C. The $R$(H₂) for IGZO621 and IGZO621:H were set at zero and 5%, respectively, while $R$(O₂) was varied from 2% to 30%. The $n_e$ of the IGZO621 films decreased with...
increasing \( R(O_2) \). An \( R(O_2) \) of more than 20% was required to reduce \( n_e \) to a value below \( 10^{18} \text{ cm}^{-3} \). In contrast, the \( n_e \) of the IGZO621:H film steeply decreased when \( R(O_2) \) increased from 2% to 5%. This result indicates that the \( R(O_2) \) value required to obtain an appropriate \( n_e \) for TFT application could be reduced by IGZO621:H. It is worth noting that the \( n_e \) of IGZO621:H is approximately one order of magnitude lower than that of IGZO621 at the same \( R(O_2) \) value. This is a phenomenon similar to that described in our previous report on IGZO111:H.\(^{17}\)

Next, the transfer characteristics of the IGZO621 and IGZO621:H TFTs were compared. Figure 2(a) shows the transfer characteristics of the TFTs with IGZO621 channels deposited at various \( R(O_2) \) values without \( H_2 \) gas measured after annealing at a \( T_a \) of 150 °C. The TFTs with \( R(O_2) \) values of 10% and 20% did not show switching properties (conductive behavior). On the other hand, the TFT with an \( R(O_2) \) of 30% showed a switching property with a threshold voltage \( (V_{th}) \) of \(-0.6 \text{ V}\) and a steep subthreshold swing (SS) of 0.09 V dec\(^{-1}\); however, a huge hysteresis \( (V_{H}) \) of 12.3 V was observed. As \( R(O_2) \) further increased to 49%, \( V_{th} \) increased to \(-18 \text{ V}\) and the on-current significantly decreased. The huge \( V_{H} \) was probably induced by oxygen ion bombardment during IGZO sputtering because weakly bonded oxygen atoms, such as excess/interstitial oxygen atoms, form electron traps in IGZO channels.\(^{24–26}\) Thus, there is a trade-off relationship between \( V_{th} \) and \( V_{H} \) in low-temperature-processed IGZO TFTs for flexible device applications.

In contrast, the TFTs with an \( R(O_2) \) of 10% exhibited excellent switching properties with a negligibly small \( V_{H} \), even after annealing at a \( T_a \) of 150 °C when IGZO621:H was used as a channel, as shown in Fig. 2(b). Moreover, \( V_{th} \) could be controlled by \( R(H_2) \) without the degradation of SS and \( V_{H} \). A \( \mu FE \) of 18.9 cm\(^2\) V\(^{-1}\) s\(^{-1}\), a \( V_{th} \) of \(-2.0 \text{ V}\), and a \( V_{H} \) of zero were achieved at \( R(O_2) = R(H_2) = 10\% – 9\% \). Figure 2(c) shows the transfer characteristics of the TFTs with IGZO621:H channels deposited at various \( R(O_2) \) values with a constant \( R(H_2) \) of 5%. By increasing \( R(O_2) \), the \( V_{th} \) of the IGZO621:H TFT could also be controlled. The obtained \( \mu FE \) of 18.7 cm\(^2\) V\(^{-1}\) s\(^{-1}\) and the \( V_{th} \) of \(-1.6 \text{ V}\) at \( R(O_2) = R(H_2) = 30\% – 5\% \) were almost the same as the values at \( R(O_2) = R(H_2) = 10\% – 9\% ; \) However, \( V_{H} \) at \( R(O_2) = R(H_2) = 30\% – 5\% \) slightly deteriorated to 0.2 V. The electrical properties of the IGZO621 and IGZO621:H TFTs are summarized in Table I.

To investigate the influences of \( R(O_2) \) and \( R(H_2) \) during IGZO sputtering on the reliability of TFTs, positive-bias temperature stress (PBTS) and negative-bias temperature stress (NBTS) tests were carried out. The stress temperature was kept at 60 °C, and the gate stress voltages for PBTS and NBTS were +20 V and −20 V, respectively. For reliability tests, a permanent epoxy-based negative photoresist (SU-8)\(^{27}\) was spin-coated on the TFTs as an organic passivation layer (OPVL) to exclude water and oxygen molecules in ambient air which degrade long-term and/or storage stability.\(^{28–30}\) Note here that an \( R(O_2) \) of 49% was used for the IGZO621 TFT with an OPVL because the TFT properties of IGZO621 with an \( R(O_2) \) of 30% vanished (conductive behavior) after the OPVL was introduced.

Figure 3 shows the changes in the transfer characteristics of the TFTs with \( R(O_2) = R(H_2) \) values of (a) 49%–0%, (b) 10%–5%, (c) 10%–9%, and (d) 30%–5% during the PBTS test. The \( V_{th} \) of the IGZO621 TFT with \( R(O_2) = R(H_2) = 49\% – 0\% \)
significantly shifted ($\Delta V_{th}$) in the positive gate voltage direction to more than 20 V right after application of PBTS for 10 s. In contrast, the reliability of the IGZO621:H TFT with $R(O_2) - R(H_2) = 10\% - 5\%$ markedly improved, and $V_{th}$ changed negligibly after application of PBTS for 10 ks.

Figure 3(e) summarizes the stress time dependence of the $\Delta V_{th}$ of the TFTs with various $R(O_2) - R(H_2)$ values during both the PBTS and NBTS tests. For the PBTS test, $\Delta V_{th}$ after applying NBTS for 10 ks was $-2.5$ V for the TFT with an $R(O_2) - R(H_2)$ value of 10\%–9\%, although the NBTS reliability also improved when the $R(O_2) - R(H_2)$ values varied from 30\%–5\% to 10\%–9\%. Several mechanisms have been proposed to explain $\Delta V_{th}$ under negative-bias stress (NBS).$^{30,31}$ It was reported that H$_2$O adsorption results in NBS degradation in IGZO TFTs with an OPVL.$^{32,33}$ In addition, the SU-8 OPVL contains a photosensitizer to initiate and catalyze cross-linking reactions during UV exposure. The photosensitizer produces an acid from which H$^+$ catalyzes the cross-linking reaction. When the cross-linking reaction is complete, some of the H$^+$ cations can bond the IGZO surface by capturing electrons; thus, hydrogen in the SU-8 or SU-8/IGZO interface would also be one of the plausible causes of the negative $\Delta V_{th}$ under NBTS.

Although further research is required to elucidate the mechanism behind the NBTS degradation, the reductions in $n_e$, and the number of electron traps in IGZO:H, the proposed method shows promise for fabricating low-temperature-processed oxide TFTs for future flexible electronics.

In summary, we investigated the electrical properties and reliability of an IGZO621:H TFT. The $n_e$ of IGZO621:H is approximately one order of magnitude lower than that of IGZO621 at the same $R(O_2)$ value after 150 °C annealing. The IGZO621:H TFT exhibited a $\mu_{FE}$ of 18.9 cm$^2$ V$^{-1}$ s$^{-1}$ and a $V_{th}$ of $-2.0$ V without hysteresis. Moreover, the reliability of the IGZO621:H TFT markedly improved as compared with that of the hydrogen-free IGZO621 TFT.

Table I. Summary of TFT properties.

| $R(O_2) - R(H_2)$ (%) | $\mu_{FE}$ (cm$^2$ V$^{-1}$ s$^{-1}$) | SS (V dec$^{-1}$) | $V_{th}$ (V) | $V_{H}$ (V) |
|------------------------|----------------------------------|-----------------|------------|------------|
| 30–0                   | 20.0                             | 0.09            | $-0.6$     | 12.3       |
| 49–0                   | 22.1                             | 0.26            | $+0.2$     | $\sim 18$ |
| 10–5                   | 18.9                             | 0.11            | $-5.8$     | 0          |
| 10–9                   | 19.3                             | 0.10            | $-2.0$     | 0          |
| 20–5                   | 20.3                             | 0.09            | $-1.7$     | 0.1        |
| 30–5                   | 18.7                             | 0.09            | $-1.6$     | 0.2        |
The $V_{th}$ of the IGZO621:H TFT changed negligibly after applying PBTS at 60 °C for 10 ks. Thus, we believe that IGZO:H will be an important channel material of low-temperature-processed TFTs for flexible electronics.

Acknowledgments Part of this research was supported by JSPS KAKENHI Grant No. 16K06309.

ORCID IDs Daichi Koretomo https://orcid.org/0000-0003-1885-0495 Yusaku Magari https://orcid.org/0000-0001-9655-4283 Mamoru Furuta https://orcid.org/0000-0003-1685-3246

1) K. Nomura, H. Ohta, A. Takagi, T. Kamiya, M. Hirano, and H. Hosono, Nature 432, 488 (2004).
2) T. Kamiya, K. Nomura, and H. Hosono, Sci. Technol. Adv. Mater. 11, 044305 (2010).
3) N. Münzenrieder, K. H. Chernenack, and G. Troster, IEEE Trans. Electron Devices 58, 2041 (2011).
4) D. Koretomo, T. Toda, T. Matsuda, M. Kimura, and M. Furuta, IEEE Trans. Electron Devices 63, 2785 (2016).
5) T. Toda, G. Tatsuoka, Y. Magari, and M. Furuta, IEEE Electron Device Lett. 37, 1006 (2016).
6) M. K. Lee, C. C. Kim, J. W. Park, E. Kim, M. L. Seol, J. Y. Park, Y. K. Choi, S. H. K. Park, and K. C. Choi, IEEE Trans. Electron Devices 64, 3179 (2017).
7) H. K. Kim, M. Furuta, and S. M. Yoon, ACS Appl. Electron. Mater. 1, 2363 (2019).
8) Y. Magari, S. G. M. Aman, D. Koretomo, K. Masuda, K. Shimpo, and M. Furuta, Jpn. J. Appl. Phys. 59, SSGJJ04 (2020).
9) T. Hiro, M. Furuta, T. Hiramatsu, T. Matsuda, C. Li, H. Furuta, H. Hokari, M. Yoshida, H. Ishii, and M. Kakagewa, IEEE Trans. Electron Devices 55, 3136 (2008).
10) Y. Hanyu, K. Abe, K. Domen, K. Nomura, H. Hiramatsu, H. Kumomi, and T. Kamiya, J. Disp. Technol. 10, 979 (2014).
11) T. Kamiya and H. Hosono, ECS Trans. 54, 103 (2013).
12) S. G. M. Aman, D. Koretomo, Y. Magari, and M. Furuta, IEEE Trans. Electron Devices 65, 3257 (2018).
13) D. Koretomo, Y. Hashimoto, S. Hamada, M. Miyanagi, and M. Furuta, Jpn. J. Appl. Phys. 58, 018003 (2018).
14) Y. J. Tak, B. D. Ahn, S. P. Park, S. J. Kim, A. R. Song, K. B. Chung, and H. J. Kim, Sci. Rep. 6, 21869 (2016).
15) W. G. Kim, Y. J. Tak, B. D. Ahn, T. S. Jung, K. B. Chung, and H. J. Kim, Sci. Rep. 6, 23039 (2016).
16) M. P. A. Jallorina, J. P. S. Bermundo, M. N. Fujii, Y. Ishikawa, and Y. Uraoka, Appl. Phys. Lett. 112, 193501 (2018).
17) S. G. M. Aman, Y. Magari, K. Shimpo, Y. Hirota, H. Makino, D. Koretomo, and M. Furuta, Appl. Phys. Express 11, 081101 (2018).
18) S. Tomai, M. Nishimura, M. Rose, M. Matsuura, K. Kasami, S. Matsuzaki, H. Kawashima, F. Utsuno, and K. Yano, Jpn. J. Appl. Phys. 51, 03CB01 (2011).
19) S. Uraoka et al., Appl. Phys. Lett. 102, 053506 (2013).
20) T. Kizu, S. Aikawa, N. Mitomo, M. Shimizu, Z. Gao, M. Li, T. Nabatame, and K. Tsukagoshi, Appl. Phys. Lett. 104, 152103 (2014).
21) M. Furuta, D. Koretomo, Y. Magari, S. G. M. Aman, R. Higashi, and S. Hamada, Jpn. J. Appl. Phys. 58, 090604 (2019).
22) Y. Hanyu, K. Domen, K. Nomura, H. Hiramatsu, H. Kumomi, and T. Kamiya, Appl. Phys. Lett. 103, 202114 (2013).
23) T. Toda, D. Wang, J. Jiang, M. Furuta, IEEE Trans. Electron Devices 61, 3762 (2014).
24) K. Ide, Y. Kikuchi, K. Nomura, M. Kimura, T. Kamiya, and H. Hosono, Appl. Phys. Lett. 99, 093507 (2011).
25) K. Ide, K. Nomura, H. Hosono, and T. Kamiya, Phys. Status Solidi A 216, 1800372 (2019).
26) S. Kim et al., IEEE Electron Device Lett. 33, 62 (2012).
27) A. Olziersky, P. Barquinha, A. Vilà, L. Pereira, G. Gonçalves, E. Fortunato, R. Martins, and J. Morante, J. Appl. Phys. 108, 064505 (2010).
28) J. S. Park, J. K. Jeong, H. J. Chung, Y. G. Mo, and H. D. Kim, Appl. Phys. Lett. 92, 072104 (2008).
29) J. K. Jeong, H. W. Yang, J. H. Jeong, Y. G. Mo, and H. D. Kim, Appl. Phys. Lett. 93, 123508 (2008).
30) J. F. Conley, IEEE Trans. Device Mater. Reliab. 10, 460 (2010).
31) Y. Uraoka, J. P. Bermundo, M. N. Fujii, M. Uenuma, and Y. Ishikawa, Jpn. J. Appl. Phys. 58, 090502 (2019).
32) J. Yang et al., Appl. Phys. Lett. 111, 073506 (2017).
33) Y. Han, C. Cui, J. Yang, M. Tsai, T. Chang, and Q. Zhang, IEEE Trans. Device Mater. Reliab. 16, 20 (2016).