J Electr Eng Technol.2017; 12(6): 2353-2358
http://doi.org/10.5370/JEET.2017.12.6.2353

ISSN(Print) 1975-0102
ISSN(Online) 2093-7423

Dynamic Response Behavior of Femtosecond Laser-Annealed Indium Zinc Oxide Thin-Film Transistors

Fei Shan* and Sung-Jin Kim†

Abstract – A femtosecond laser pre-annealing process based on indium zinc oxide (IZO) thin-film transistors (TFTs) is fabricated. We demonstrate a stable pre-annealing process to analyze surface structure change of thin films, and we maintain electrical stability and improve electrical performance. Furthermore, dynamic electrical characteristics of the IZO TFTs were investigated. Femtosecond laser pre-annealing process–based IZO TFTs exhibit a field-effect mobility of 3.75 cm^2/Vs, an I_{on}/I_{off} ratio of 1.77×10^5, a threshold voltage of 1.13 V, and a subthreshold swing of 1.21 V/dec. And the IZO-based inverter shows a fast switching behavior response. From this study, IZO TFTs from using the femtosecond laser annealing technique were found to strongly affect the electrical performance and charge transport dynamics in electronic devices.

Keywords: IZO thin-film transistor, Femtosecond laser, Dynamic response, Pre-annealing, Solution process

1. Introduction

Thin-film transistors (TFTs) [1], [2] have been researched extensively as switching devices in active matrix liquid crystal displays (AM-LCDs) [3], organic light emitting diodes (OLEDs) [4], and flexible displays [5], resulting in enormous demand for models that can predict the switching devices' dynamic behavior with optical characteristics. Increasingly stringent requirements on the dynamic response of the TFT backplane technology for AM-LCDs, OLEDs, and flexible displays undoubtedly contribute to improvements in display resolution and frame rate, also further reducing charging time of each pixel. The dynamic measurements are crucial for design optimization of AM-LCDs, OLEDs, and flexible displays.

Recently, a number of studies have explored solution-processing of indium zinc oxide (IZO) [6-8] TFTs owing to their high electron mobility, high electrical uniformity over a large substrate size, and low processing temperature.

In particular, the IZO TFT has a much higher current driving performance than a-Si:H TFTs [9, 10], so it is viewed as a valuable thin film–device candidate for next-generation ultra-high–resolution flat-panel displays. Although high-performance oxide TFTs have previously been reported, solution-processed IZO TFTs using a single near-infrared femtosecond laser pulse are still rare. With a femtosecond laser pre-annealing process [11-13], permanent gratings have been encoded on the surface of a silicon dioxide (SiO_2) thin film. Femtosecond laser technology is applied in most research areas of thin-film devices. The laser annealing process shows special advantages: electric performance improvement and a fast and stable preparation process compared with the traditional preparation methods as vacuum furnace, ultraviolet, and so on. In addition, an extremely short laser heat treatment time also plays a role in reducing the deterioration of an integrated circuit and ensuring its electrical properties.

In this paper, our objective is to compare and analyze the electrical performance of general IZO TFTs and femtosecond laser–annealed IZO TFTs with a laser heat treatment time of 5 s; meanwhile, the dynamic response of femtosecond laser–annealed IZO TFTs was investigated.

2. Experiments

A precursor solution for fabricating IZO thin films was prepared by dissolving 0.1 M of indium nitrate hydrate [$In(NO_3)_3 \cdot xH_2O$] and 0.1 M of zinc acetate dehydrate [$Zn(CH_3COO)_2 \cdot 2H_2O$]. First, we added 93 mg of indium powder into a reagent bottle, which was then mixed with 2.5 ml of 2-methoxyethanol, 50 μl of acetylacetone, and 22.5 μl of NH_3 to form a stable indium nitrate hydrate solution. And in a similar way, we formed a zinc acetate dehydrate solution with 33 mg of zinc powder, 1.5 ml of 2-methoxyethanol, and 30 μl of acetylacetone. To facilitate the dissolving process, the indium nitrate hydrate solution and zinc acetate dehydrate solution were stirred at 60 °C and 700 rpm for 1 hour with a magnetic stirrer. Then, an IZO solution was made from 2.1 ml of an indium nitrate hydrate solution and 0.9 ml of a zinc acetate dehydrate solution with a molar ratio of 7:3, and it was stirred vigorously at 27 °C and 500 rpm for 2 hours to form a transparent IZO solution.

† Corresponding Author: College of Electrical and Computer Engineering, Chungbuk National University, Korea. (ksj@cbnu.ac.kr)
* College of Electrical and Computer Engineering, Chungbuk National University, Korea. (my521241sf@hotmail.com)
Received: May 15, 2017; Accepted: August 5, 2017

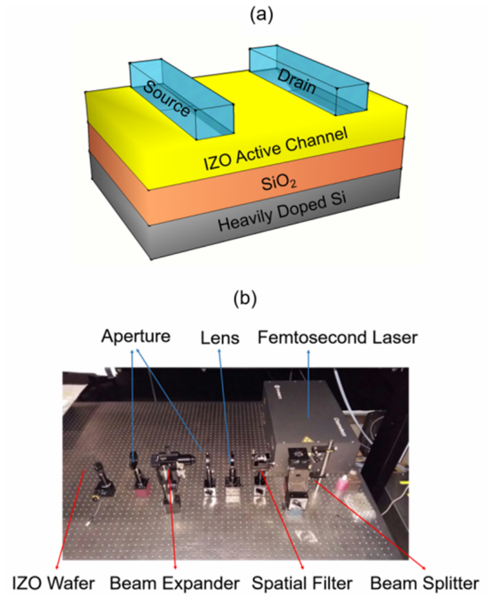


Fig. 1. (a) Schematic representation of the IZO-based TFT structure, and (b) a simplified schematic of the setup for the optical experiment

A heavily doped–type (n++) silicon substrate was used in a top source/drain, bottom gate contact structure for the fabrication of IZO TFTs, as shown in Fig. 1 (a), and 100 nm thick silicon dioxide was thermally grown on top of the substrate as a gate dielectric. The IZO solution was spin-coated on the SiO_2 substrate at 3000 rpm for 30 s as a 30 nm IZO thin film. The channel length, L, and width, W, of the TFTs were 200 µm and 2000 µm, respectively. A simplified femtosecond laser pre-annealing scheme works as shown in Fig. 1(b). The femtosecond laser system (Coherent, Chameleon Ultra II) has a center wavelength of 800 nm, a pulse width of 140 fs, and average power of 3 W/cm^2. A suitable femtosecond laser pulse was separated out via beam splitter, then the pulse was expanded using an objected lens embedded in a spatial filter, using a beam expander to ensure the IZO substrate was entirely covered by the femtosecond laser pulse. The substrate had been heat-treated via femtosecond laser for 5 s, and then annealed in a vacuum furnace at 380 °C for 2 h. And then, the top-contact source and drain electrodes were deposited on top of the IZO layer via metal evaporator using 100 nm thick aluminum [Al]. Meanwhile, we had fabricated another IZO substrate without using a femtosecond laser pulse to conduct a contrast test. The electrical characteristics were measured with an Agilent 1500B semiconductor parameter analyzer at room temperature in the dark, and the surface structure properties were investigated by atomic force microscopy (AFM).

3. Discussion

The surface of the active layer had a lot of bond energy

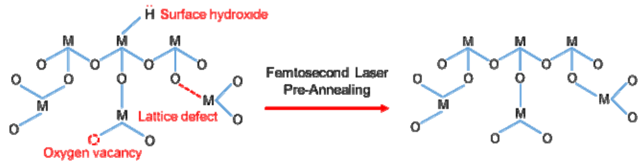


Fig. 2. Schematics for the process of removing with femtosecond laser pre-annealing

of diatomic formations (In-O and Zn-O), which were determined by chemical bond strength and were defined as the minimum energy required to dissociate a diatomic species into single atoms [14-16]. The diatomic In-O and Zn-O in the IZO active layer can be broken when the actual external energy exceeds the bond energy by femtosecond laser irradiation. The femtosecond laser irradiation energy can be calculated as Eq. (1).

$$E = hc/\lambda \quad (1)$$

where E-energy, J/cm^2; h-Planck's constant, J/s; c-speed of light, m/s; λ-wavelength. By the femtosecond laser irradiation at a power of 3 W/cm^2 and wavelength of 800 nm, the energy was sufficient to break the chemical bonds. And activation energy increased with the increase of oxygen partial pressure to urge atomic reorganization. We found that using femtosecond laser pulse heat treatment on the thin films achieved the improvement. First, the diatomic In-O and Zn-O can be broken by femtosecond laser pulse irradiation of higher energy than bond energy. This laser heat treatment process converted In-O and Zn-O to metal-oxygen-network bonding as In-O-Zn-O or Zn-O-In-O [17]. And using the femtosecond laser pre-annealing process with exceedingly brief heat treatment at 5 s could effectively avoid generating excessive temperatures, which led to the oxygen vacancy. Meanwhile, the decomposition rate of surface hydroxide increased with the increase in surface temperature, and a considerable amount of hydroxide was decomposed with the femtosecond laser pre-annealing process. It can be seen that the lattice defect of varying dimensions and deep rill-like folds had been repaired. Therefore, femtosecond laser heat treatment improved the quality of the thin films because of a reduction in the defect-related oxygen vacancies and an increase in the chemical bonds, as shown in Fig. 2. The AFM images clearly exhibit some changes on the surface. It can be seen that the surface shows more homogeneous deep rill-like folds, as shown in Fig. 3(b). The thin film with femtosecond laser heat treatment has a smooth surface with a root mean square roughness of about 1.92 nm.

Fig. 4 (a) shows the output curves (I_d-V_d) of the n-type TFTs with a 30 nm thick IZO active layer from various annealing processes, exhibiting similar linearity of the drain current (I_d) in the low-V_d region [18], but the linearity with femtosecond laser shows good saturation in the high-V_d region. Fig. 4 (b) shows the transfer curves of the

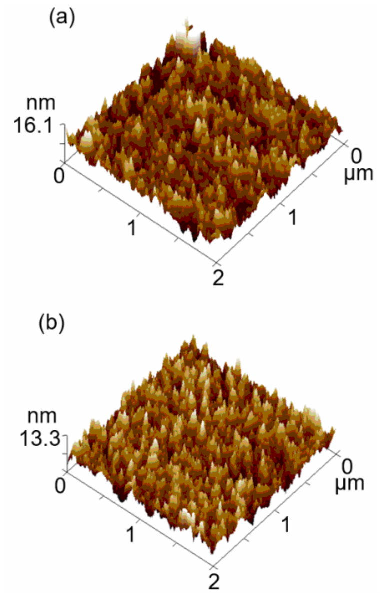


Fig. 3. AFM 3D topographies: (a) without femtosecond laser pre-annealing, and (b) with femtosecond laser pre-annealing at 5 s

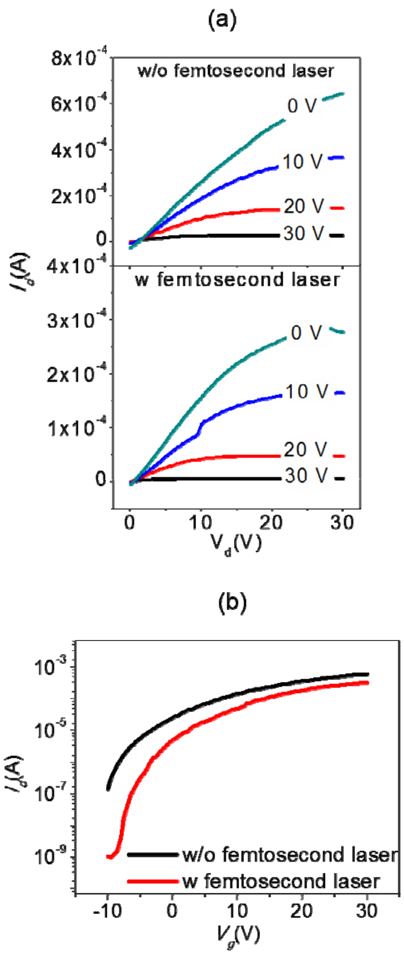


Fig. 4. Contrastive analysis of IZO TFTs without (w/o) and with (w) femtosecond laser pre-annealing of 5 s on (a) output characteristics, and (b) transfer curves

IZO TFTs with a drain-source voltage (V_d) of 25 V. On current (I_{on}) is greatly improved with femtosecond laser pre-annealing compared to without the laser pulse. This is because interface traps and lattice defects can capture electrons and holes near the interface, and minority positive-donors with oxygen vacancies are trapped at the interface. All these prevent the electrons and holes from continuing their steady drift, and reduce carrier mobility. However, short-time femtosecond laser heat treatment easily reduces channel interface traps, mobile ions, and oxygen vacancies in order to improve the electrical performance. Electrical parameters such as field-effect mobility and threshold voltage are determined from a plot of $I_d^{1/2}$ vs V_g on the basis of the following relationship in the saturation region. In addition, the subthreshold slope (S) is calculated using the following formula as Eqs. (2-3).

$$I_d = \frac{WC_i}{2L}\mu\left(V_g - V_{th}\right)^2 \tag{2}$$

$$S = \left[\left.\frac{d\log\left(I_{DS}\right)}{dV_g}\right|max\right]^{-1} \tag{3}$$

Here, W, L, μ, C_i, and V_{th} are the channel width, channel length, field-effect mobility, capacitance per unit area of the SiO_2 gate insulator (dielectric constant ~3.9), and the threshold voltage, respectively, and V_{th} is derived from the linear fit with the abscissa. IZO TFTs with femtosecond laser pre-annealing at 5 s show high field-effect mobility (μ) of 3.75 cm^2/Vs, an I_{on}/I_{off} ratio of 1.77×10^5, a threshold voltage of 1.13 V, and a subthreshold swing (S) of 1.21 V/dec, as shown in Table 1. Excess electric charge has an impact on V_{th} on an interface between an oxide and a silicon surface. Trapped carrier (electrons and holes) and structure defect may exist in this electric charge. By a short femtosecond laser pretreatment process, the electron-holes which were captured by interface traps near the interface could recombine and continue their steady motion as quasi-free particles. Meanwhile, the laser heat treatment process could optimize the structure defect, and enable the IZO layer to thinner and more homogeneous as shown in Fig. 3. A non-linear relationship between carrier concentration and electrical resistivity of sample is closely related with the oxygen vacancy. The high-temperature (380°C) vacuum furnace annealing process could result in overmuch oxygen vacancy, usually causing the increased carrier concentration

Table 1. Electrical characteristics contrasted in IZO TFTs without femtosecond laser pre-annealing and with femtosecond laser pre-annealing at 5 s

w/o and w femtosecond laser pre-annealing	μ (cm^2/Vs)	I_{on}/I_{off}	V_{th} (V)	S (V/dec)
w/o	3.50	3.03×10^3	-5.79	2.45
w	1.32 A	1.09 V	1.13	1.21

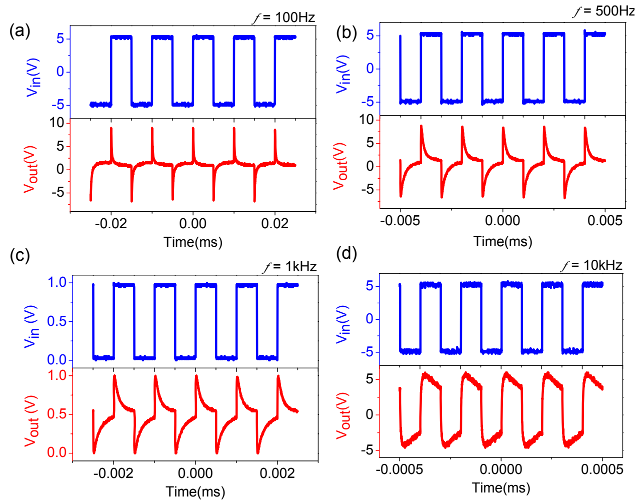


Fig. 5. Dynamic response of an n-channel inverter with load resistance R = 1 MΩ at different frequencies: (a) 100 Hz, (b) 500 Hz, (c) 1 kHz, and (d) 10 kHz

and reduced electrical resistivity. This could ultimately lead to higher leakage current. But we could obtain more stable IZO layer by the femtosecond laser pretreatment process to reduce the oxygen vacancy.

The femtosecond laser–treated inverter with depletion load demonstrates excellent dynamic performance. The transient electrical response of the inverter was investigated for *n*-channel–based inverters with load resistance R=1 MΩ at different input pulse frequencies. In order to research the dynamic response behavior of the *n*-channel TFT [19-21], we manufactured a simple load-type inverter. Fig. 5(a) shows the inverter characteristics when the square wave input bias (between –5 V to +5 V) was applied at 100 Hz and when the output voltage was switched from on to off. We measured the switching response time of resistor-loaded inverters with a 100 Hz input voltage to research the relationship between electrical performance and dynamic response. Rising time (T_r) and falling time (T_f) are defined as the times required to increase from 10% to 90% of the output level or to fall from 90% to 10% [22-24]. The entire output signal exhibited decent switching behavior with different response times, and obtained normal T_r and T_f of the inverter [25]. But T_r and T_f become shorter from increasing the frequency to 500 Hz, 1 kHz, and 10 kHz, as shown in Figs. 5(b), (c) and (d). The results with the inverter imply that IZO TFTs in conjunction with the femtosecond laser pre-annealing process can be successfully applied to electronic logic circuits.

4. Conclusion

In summary, we demonstrated IZO TFTs using a femtosecond laser pre-annealing process. We showed that the IZO TFT preparation method with femtosecond laser pre-annealing could improve electrical properties, and the dynamic electrical characteristics of the n-channel IZO TFT were examined. The short-time laser annealing process (laser irradiation at 5 s) exhibits a field-effect mobility of 3.75 cm^2/Vs, an I_{on}/I_{off} ratio of 1.77×10^5, a threshold voltage of 1.13 V, and a subthreshold swing of 1.21 V/dec. And the TFT shows good static and fast response speed. The developed femtosecond laser pre-annealing technique is promising for metal oxide semiconductor electronics.

Acknowledgements

This research was supported by the Basic Science Research Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Education (2017R1D1A3B03029782) and by a Human Resources Development, Korea Institute of Energy Technology Evaluation and Planning (KETEP) grant funded by the Korea government Ministry of Trade, industry & Energy (No. 20144030200450). This research was also supported by the Ministry of Science, ICT and Future Planning (MSIP), Korea, under the Information Technology Research Center (ITRC) support program (IITP-2017-2015-0-00448) supervised by the Institute for Information & communications Technology Promotion (IITP).

Reference

[1] K. Nomura, H. Ohta, K. Ueda, T. Kamiya, M. Hirano, and H. Hosono, "Thin-Film Transistor Fabricated in Single-Crystalline Transparent Oxide Semiconductor," *Science,* vol. 300, no. 5623, pp. 1269-1272, May 2003.

[2] M. Sato and A. J. Sievers, "Direct observation of the discrete character of intrinsic localized modes in an antiferromagnet," *Nature,* vol. 432, no. 7016, pp.486-488, Nov. 2004.

[3] J. F. Wager, B. Yeh, R. L. Hoffman, and D. A. Keszler, "An amorphous oxide semiconductor thin-film transistor route to oxide electronics," *Curr Opin Solid St M,* vol. 18, no. 2, pp. 53-61, Apr. 2014.

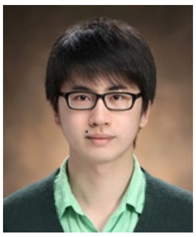
[4] H. Sasabe and J. Kido, "Development of high performance OLEDs for general lighting," *J Mater Chem C,* vol. 1, no. 9, pp. 1699-1707, Jan. 2013.

[5] J. S. Park, T. W. Kim, D. Stryakhilev, J. S. Lee, S. G. An, Y. S. Pyo, D. B. Lee, Y. G. Mo, D. U. Jin, and H. K. Chung, "Flexible full color organic light-emitting diode display on polyimide plastic substrate driven by amorphous indium gallium zinc oxide thin-film transistors," *Appl Phys Lett,* vol. 95, no. 1, p. 013503, June 2009.

[6] D. H. Lee, Y. J. Chang, G. S. Herman, and C. H. Chang, "A General Route to Printable High-Mobility Transparent Amorphous Oxide Semiconductors," *Adv Mater,* vol. 19, no. 6, pp. 843-847, Feb. 2007.

[7] H.-W. Park, K. Park, J.-Y. Kwon, D. Choi, and K.-B.

Chung, "Effect of Active Layer Thickness on Device Performance of Tungsten-Doped InZnO Thin-Film Transistor," *IEEE T Electron Dev,* vol. 64, no. 1, pp. 159-163, Jan. 2017.
[8] B. Yaglioglu, H. Y. Yeom, R. Beresford, and D. C. Paine, "High-mobility amorphous In2O3-10wt%ZnO thin film transistors," *Appl Phys Lett,* vol. 89, no. 6, p. 062103, June 2006.
[9] G. J. Lee, J. Kim, J.-H. Kim, S. M. Jeong, J. E. Jang, and J. Jeong, "High performance, transparent a-IGZO TFTs on a flexible thin glass substrate," *Semicond Sci Tech,* vol. 29, no. 3, p. 035003, Jan. 2014.
[10] S. Morawiec, M. J. Mendes, S. A. Filonovich, T. Mateus, S. Mirabella, H. Águas, I. Ferreira, F. Simone, E. Fortunato, and R. Martins, "Broadband photocurrent enhancement in a-Si: H solar cells with plasmonic back reflectors," *Opt Express*, vol. 22, no. 104, pp. A1059-A1070, June 2014.
[11] L. Hong, X. Wang, H. Zheng, H. Wang, and H. Yu, "Femtosecond laser fabrication of large-area periodic surface ripple structure on Si substrate," *Appl Surf Sci,* vol. 297, no., pp. 134-138, Apr. 2014.
[12] C. Lee, P. Srisungsitthisunti, S. Park, S. Kim, X. Xu, K. Roy, D. B. Janes, C. Zhou, S. Ju, and M. Qi, "Control of current saturation and threshold voltage shift in indium oxide nanowire transistors with femtosecond laser annealing," *Acs Nano,* vol. 5, no. 2, pp. 1095-1101, Jan. 2011.
[13] J.-M. Shieh, Z.-H. Chen, B.-T. Dai, Y.-C. Wang, A. Zaitsev, and C.-L. Pan, "Near-infrared femtosecond laser-induced crystallization of amorphous silicon," *Appl Phys Lett,* vol. 85, no. 7, pp. 1232-1234, June 2004.
[14] Y. J. Tak, D. H. Yoon, S. Yoon, U. H. Choi, M. M. Sabri, B. D. Ahn, and H. J. Kim, "Enhanced Electrical Characteristics and Stability Via Simultaneous Ultraviolet and Thermal Treatment of Passivated Amorphous In–Ga–Zn–O Thin-Film Transistors," *Acs Appl Mater Inter,* vol. 6, no. 9, pp. 6399-6405, Apr. 2014.
[15] J. M. Kwon, J. Jung, Y. S. Rim, D. L. Kim, and H. J. Kim, "Improvement in negative bias stress stability of solution-processed amorphous In–Ga–Zn–O thin-film transistors using hydrogen peroxide," *Acs Appl Mater Inter,* vol. 6, no. 5, pp. 3371-3377, Feb. 2014.
[16] Y.-H. Kim, J.-S. Heo, T.-H. Kim, S. Park, M.-H. Yoon, J. Kim, M. S. Oh, G.-R. Yi, Y.-Y. Noh, and S. K. Park, "Flexible metal-oxide devices made by room-temperature photochemical activation of sol-gel films," *Nature,* vol. 489, no. 7414, pp. 128-132, Sep. 2012.
[17] D. J. Kim, D. L. Kim, Y. S. Rim, C. H. Kim, W. H. Jeong, H. S. Lim, and H. J. Kim, "Improved electrical performance of an oxide thin-film transistor having multistacked active layers using a solution process," *Acs Appl Mater Inter,* vol. 4, no. 8, pp. 4001-4005, July 2012.
[18] Y. Wang, X. W. Sun, G. K. L. Goh, H. V. Demir, and H. Y. Yu, "Influence of channel layer thickness on the electrical performances of inkjet-printed In-Ga-Zn oxide thin-film transistors," *IEEE T Electron Dev,* vol. 58, no. 2, pp. 480-485, Feb. 2011.
[19] G. E. Park, J. Shin, D. H. Lee, T. W. Lee, H. Shim, M. J. Cho, S. Pyo, and D. H. Choi, "Acene-Containing Donor–Acceptor Conjugated Polymers: Correlation between the Structure of Donor Moiety, Charge Carrier Mobility, and Charge Transport Dynamics in Electronic Devices," *Macromolecules,* vol. 47, no. 11, pp. 3747-3754, May 2014.
[20] M. Jea, A. Kumar, H. Cho, D. Yang, H. Shim, A. K. Palai, and S. Pyo, "An organic microcrystal array-embedded layer: highly directional alternating p-and n-channels for ambipolar transistors and inverters," *J Mater Chem C*, vol. 2, no. 20, pp. 3980-3987, Feb. 2014.
[21] X. Huang, C. Wu, H. Lu, F. Ren, D. Chen, Y. Liu, G. Yu, R. Zhang, Y. Zheng, and Y. Wang, "Large-swing a-IGZO inverter with a depletion load induced by laser annealing," *IEEE Electr Device L,* vol. 35, no. 10, pp.1034-1036, Aug. 2014.
[22] H. Na, H. Cho, K. Sim, H. Shim, S.-J. Kim, and S. Pyo, "Electrical responses of short-channel organic transistor prepared by solution-processed organic crystal wire mask," *Org Electron,* vol. 15, no. 11, pp. 2728-2733, Nov. 2014.
[23] J. Kwon, H. Na, A. K. Palai, A. Kumar, U. Jeong, S. Cho, and S. Pyo, "Utilization of simply alkylated diketopyrrolopyrrole derivative as a p-channel semiconductor for organic devices," *Synthetic Met,* vol. 209, no., pp. 240-246, Nov. 2015.
[24] J. Y. Kim, D. S. Yang, J. Shin, D. Bilby, K. Chung, H. A. Um, J. Chun, S. Pyo, M. J. Cho, and J. Kim, "High-Performing Thin-Film Transistors in Large Spherulites of Conjugated Polymer Formed by Epitaxial Growth on Removable Organic Crystalline Templates," *Acs Appl Mater Inter,* vol. 7, no. 24, pp. 13431-13439, June 2015.
[25] G. Horowitz, M. E. Hajlaoui, and R. Hajlaoui, "Temperature and gate voltage dependence of hole mobility in polycrystalline oligothiophene thin film transistors," *J Appl Phys*, vol. 87, no. 9, pp. 4456-4463, Apr. 2000.



Fei Shan received the B.S. and M.S. degrees from the Department of Computer Engineering, Chungbuk National University, Cheongju, Korea, in 2015 and 2017, respect-tively. He is currently pursuing the Ph.D. degree at the Graduate School of the Chungbuk

National University, Korea. His current research interests include the fabrication of soluble oxide transistors and next generation memory devices.

Sung-Jin Kim received the B.S. and M.S. degrees from the Department of Electrical and Electronics Engineering, Kyungpook National University, Daegu, Korea, in 1999 and 2001, respectively, and the Ph.D. degree from the School of Electrical and Computer Engineering, Seoul National University, Seoul, Korea, in 2006. He worked on Samsung SDI Company, Suwon, Korea, from 2004 to 2007, the Cooperated Research and Development Center and developed various next-generation displays like flexible/bendable light sources and active matrix organic light-emitting diodes as a Senior Research Engineer. In 2007, he was a Post-Doctoral Research Scientist with the Department of Electrical Engineering, Columbia University, New York, NY, USA, where he was initially engaged in research on the application of organic thin-film transistors and new processing strategies for highly integrated organic systems. In 2008, he joined the School of Electrical and Computer Engineering, Georgia Institute of Technology, Atlanta, GA, USA, as a Post-Doctoral Fellow working on solution-processable organic light emitting diodes. In 2010, he was with the College of Electrical and Computer Engineering, Chungbuk National University, Cheongju, Korea, as an Assistant Professor. His current research interests include the organic/oxide devices, flexible printing electronics, large-area light sources, and energy harvesting applications.