

# Small-Subthreshold-Swing and Low-Voltage Flexible Organic Thin-Film Transistors Which Use HfLaO as the Gate Dielectric

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**Abstract**—Pentacene organic thin-film transistors (OTFTs) with a high- $\kappa$  HfLaO dielectric were integrated onto flexible polyimide substrates. The pentacene OTFTs exhibited good performance, such as a low subthreshold swing of 0.13 V/decade and a threshold voltage of  $-1.25$  V. The field-effect mobility was  $0.13 \text{ cm}^2/\text{V} \cdot \text{s}$  at an operating voltage as low as only 2.5 V. These characteristics are attractive for high-switching-speed and low-power applications.

**Index Terms**—Flexible, HfLaO, high- $\kappa$ , organic thin-film transistors (OTFTs), pentacene.

## I. INTRODUCTION

PENTACENE-BASED organic thin-film transistors (OTFTs) have been intensely investigated due to their low cost and lightweight for potential use in applications such as flexible displays and low-cost flexible integrated circuits (ICs) [1]–[3]. The low thermal budget and rapid processing have strong merits of energy savings and environment friendliness, which are in sharp contrast to the extended 600-°C annealing times in conventional solid-phase-crystallized (SPC) poly-Si TFTs. Although low-thermal-budget poly-Si TFTs can also be formed on plastic substrate using excimer laser annealing [4], the uniformity is a concern. Alternatively, poly-Si TFTs [5] or even single-crystal submicrometer MOSFETs [6] can be realized on plastic substrate by fabrication first, separation next and transfer, but these methods still require high thermal budget for device fabrication. However, conventional OTFTs require a high operating voltage and show a poor subthreshold swing (SS), which detracts from their suitability in IC operations [7], [8]. To address these issues, high- $\kappa$  gate dielectrics have been applied in OTFTs for low-voltage operation [1], [3],

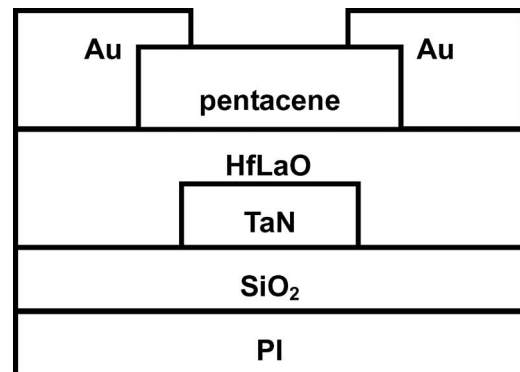
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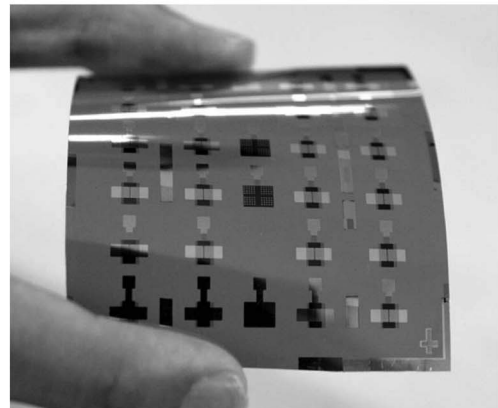
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(a)



(b)

Fig. 1. Schematic diagram (a) and image (b) of the high- $\kappa$  flexible HfLaO/pentacene OTFTs.

[9]–[13]. We have previously reported pentacene OTFTs on  $\text{SiO}_2/\text{Si}$  substrates using high- $\kappa$  HfLaO as the gate dielectric [13]. Although the performance is comparable with that of SPC poly-Si TFTs, the process temperature of 350 °C is still not suitable for flexible electronics. In this letter, we further decrease the process temperature to 200 °C and demonstrate HfLaO/pentacene OTFTs, fabricated on low-cost flexible polyimide (PI) (Kapton HPP-ST, Dupont) substrates. These substrates are much more economical than other PI (Kapton E-type, Dupont) substrates [1], [9] and those which use polyethylene naphthalate (Teonex Q65 PEN, Dupont) [2]. Our HfLaO/pentacene OTFTs showed a low SS of only 0.13 V/decade, a high gate capacitance density of  $450 \text{ nF}/\text{cm}^2$ ,

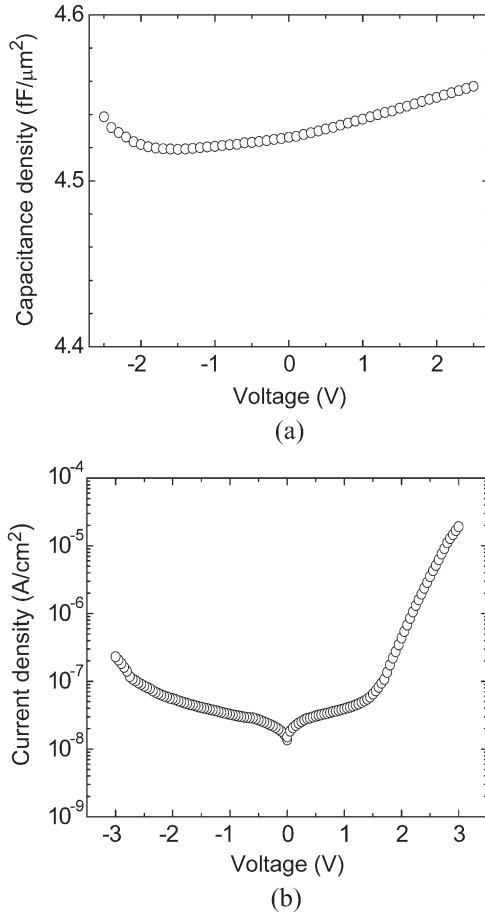


Fig. 2. (a)  $C$ - $V$  and (b)  $J$ - $V$  characteristics of Au/HfLaO/TaN capacitors.

a low threshold voltage ( $V_T$ ) of  $-1.25$  V, a good mobility ( $\mu_{FE}$ ) of  $0.13$   $\text{cm}^2/\text{V}\cdot\text{s}$ , and an ON-OFF-state drive current ratio ( $I_{on}/I_{off}$ ) of  $1.2 \times 10^4$ . This superior performance permits the devices to be operated at  $2.5$  V, which could be useful in flexible electronics.

## II. EXPERIMENTAL DETAILS

All the devices were fabricated on  $125\text{-}\mu\text{m}$ -thick PI substrates (Kapton HPP-ST, Dupont). Prior to the device fabrication process, the PI substrates were annealed in vacuum at  $200^\circ\text{C}$  [2]. A  $100\text{-nm}$   $\text{SiO}_2$  thin film was deposited on the PI substrate [3]. Then, a  $50\text{-nm}$  TaN gate electrode was deposited by reactive sputtering through a shadow mask. The surface of the TaN gate was treated in a  $\text{NH}_3$  plasma to reduce the gate leakage current [14], [15]. A  $30\text{-nm}$ -thick HfLaO gate dielectric was then deposited. A  $200^\circ\text{C}$   $30\text{-min}$  furnace  $\text{O}_2$  treatment was then used to improve the gate oxide quality. Next, the pentacene active layer (Aldrich Chemical Company),  $70$  nm in thickness, was deposited through the shadow mask under vacuum and  $70^\circ\text{C}$ . Finally,  $50$  nm of Au was deposited for the source/drain electrodes. The channel width and channel length were  $2000$  and  $100$   $\mu\text{m}$ , respectively. Metal-insulator-metal Au/HfLaO/TaN capacitors,  $200 \times 200$   $\mu\text{m}^2$  in size, were also fabricated to analyze the leakage current and the dielectric properties.

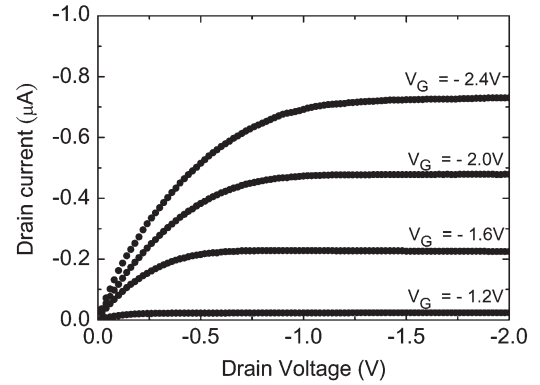


Fig. 3.  $I_D$ - $V_D$  curve for a HfLaO gate dielectric OTFT.

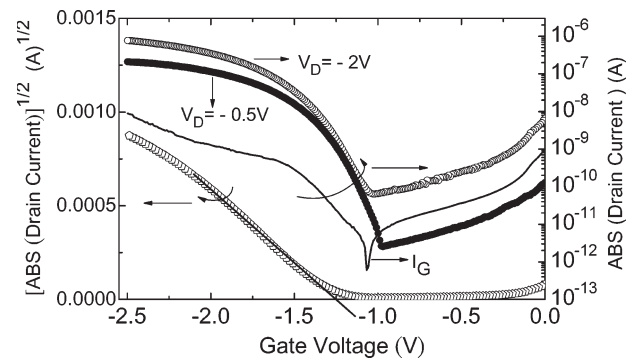


Fig. 4.  $I_D$ - $V_G$  and  $-I_D^{1/2}V_G$  of a HfLaO gate dielectric OTFT.

## III. RESULTS AND DISCUSSION

We show a schematic diagram and an image of the OTFTs in Fig. 1(a) and (b), respectively. The  $C$ - $V$  and  $J$ - $V$  characteristics of the Au/HfLaO/TaN capacitors are shown in Fig. 2(a) and (b), respectively. A low leakage current of  $3.5 \times 10^{-6}$   $\text{A}/\text{cm}^2$  at  $2.5$  V was measured, along with a capacitance density of  $450$   $\text{nF}/\text{cm}^2$ . This density yields an equivalent oxide thickness (EOT) of  $7.7$  nm and a high- $\kappa$  value of  $15.3$ .

The output characteristics ( $I_D$ - $V_D$ ) of a high- $\kappa$  HfLaO OTFT are shown in Fig. 3. Good drain saturation behaviors were observed and suggest possible operation at  $2.5$  V. Fig. 4 shows the  $I_D$ - $V_G$  characteristics of a representative OTFT, and  $\mu_{FE}$  and  $V_T$  were determined from the  $-I_D^{1/2}$  versus  $V_G$  plot. The resulting values are  $-1.25$  V for  $V_T$ ,  $0.13$   $\text{cm}^2/\text{Vs}$  for  $\mu_{FE}$ , and an SS of  $0.13$  V/decade. The  $I_{on}/I_{off}$  ratio was  $1.2 \times 10^4$ . The SS of our device is better than the values observed for other flexible pentacene OTFTs [1]-[3], [9], [10], arising from the high gate capacitance density and small EOT [13]. The relatively smaller mobility and  $I_{on}/I_{off}$  were due to both lower operation voltage and poor surface roughness. An rms surface roughness of  $4.3$  nm was measured by atomic force microscopy on HfLaO and worse than the  $2.0\text{-nm}$  value of BZN [3], which originates from the poor  $9.0\text{-nm}$  roughness of very low cost PI substrate (Kapton HPP-ST, Dupont).

In Table I, we summarize some important device parameters, including other data of low-voltage flexible pentacene OTFTs using  $\text{Bi}_{1.5}\text{Zn}_{1.0}\text{Nb}_{1.5}\text{O}_7$  (BZN), polyvinylphenol,  $\text{Ta}_2\text{O}_5$ ,  $\text{TiSiO}_2$ , and Mn-doped  $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$  (Mn-doped BST) as

TABLE I  
COMPARISON OF LOW-VOLTAGE OTFTs WITH VARIOUS GATE DIELECTRICS

OTFTs with various gate dielectrics	HfLaO [This work]	BZN [1]	PVP [2]	Ta <sub>2</sub> O <sub>5</sub> [3]	Mn-doped BST [9]	TiSiO <sub>2</sub> [10]	$\pi$ - $\sigma$ -PA1 /AlO <sub>x</sub> [11]	OPDA/ AlO <sub>x</sub> [12]	HfLaO [13]
substrate	PI Kapton HPP-ST	PI Kapton E-type	PEN	PEN	PI Kapton E-type	PEN	Si	glass	SiO <sub>2</sub> /Si
Dielectric thickness (nm)	30	200	50	150	200	136.4	-	-	20
Operating voltage (V)	2.5	2	8	12	10	5	3	2	2
C <sub>i</sub> (nF/cm <sup>2</sup> )	450	221	63.7	141.6	110	142.1	760	600-800	950
V <sub>T</sub> (V)	-1.25	0.1	-	0.8	-1	-0.88	-1.3	0.35	-1.3
$\mu_{FE}$ (cm <sup>2</sup> /Vs)	0.13	0.5	0.1	0.25	0.32	0.67	0.18	0.04	0.71
SS (V/decade)	0.13	0.3	0.6	-	-	0.315	0.085	0.11	0.078
$\mu_{FE}C_i$ (nF/cm <sup>2</sup> )	58.5	110.5	6.37	35.4	35.2	95.21	136.8	24-32	674.5
I <sub>on</sub> /I <sub>off</sub>	1.2×10 <sup>4</sup>	3.5×10 <sup>5</sup>	1×10 <sup>4</sup>	-	<1×10 <sup>3</sup>	1×10 <sup>4</sup>	1×10 <sup>5</sup>	1×10 <sup>4</sup>	1×10 <sup>5</sup>

gate dielectrics, and fabricated on high-quality PI (Kapton E-type) and PEN substrates [1]–[3], [9], [10]. The low-voltage OTFTs using (2-anthryl)undecoxycarbonyldecylphosphonic acid ( $\pi$ - $\sigma$ -PA1)/AlO<sub>x</sub>, octadecylphosphonic acid (OPDA)/AlO<sub>x</sub>, and HfLaO as gate dielectrics, fabricated on Si, glass, and SiO<sub>2</sub>/Si substrate, are also listed in Table I for comparison [9]–[13]. The value of the  $\mu_{FE}C_i$  term is directly proportional to  $I_D (W/2L_G \times \mu_{FE}C_i(V_G - V_T)^2)$ , normalized to the channel length  $L_G$ , channel width  $W$ , and overdrive voltage of  $V_G - V_T$ . The performance of our HfLaO OTFTs is comparable with that of other low-voltage flexible pentacene OTFTs for low-power applications, but with the additional merit of a good SS and the use of economical PI substrates.

#### IV. CONCLUSION

A high- $\kappa$  HfLaO dielectric was successfully integrated into pentacene OTFTs fabricated on low-cost flexible substrates. The HfLaO/pentacene OTFTs showed a low SS, a small  $V_T$ , a large  $\mu_{FE}C_i$ , and a low operating voltage.

#### REFERENCES

- [1] Y. W. Choi, I.-D. Kim, H. L. Tuller, and A. I. Akinwande, "Low-voltage organic transistors and depletion-load inverters with high- $K$  pyrochlore BZN gate dielectric on polymer substrate," *IEEE Trans. Electron Devices*, vol. 52, no. 12, pp. 2819–2824, Dec. 2005.
- [2] H. Klauk, M. Halik, U. Zschieschang, F. Eder, D. Rohde, G. Schmid, and C. Dehm, "Flexible organic complementary circuits," *IEEE Trans. Electron Devices*, vol. 52, no. 4, pp. 618–622, Apr. 2005.
- [3] M. Mizukami, N. Hirohata, T. Iseki, K. Ohtawara, T. Tada, S. Yagyū, T. Abe, T. Suzuki, Y. Fujisaki, Y. Inoue, S. Tokito, and T. Kurita, "Flexible AM OLED panel driven by bottom-contact OTFTs," *IEEE Electron Device Lett.*, vol. 27, no. 4, pp. 249–251, Apr. 2006.
- [4] D. P. Gosan, T. Noguchi, and S. Usui, "High mobility thin film transistors fabricated on a plastic substrate at a processing temperature of 110 °C," *Jpn. J. Appl. Phys.*, vol. 39, pp. L179–L181, 2000.
- [5] Y. Lee, H. Li, and S. J. Fonash, "High-performance poly-Si TFTs on plastic substrates using a nano-structured separation layer approach," *IEEE Electron Device Lett.*, vol. 24, no. 1, pp. 19–21, Jan. 2003.
- [6] H. L. Kao, A. Chin, B. F. Hung, J. M. Lai, C. F. Lee, M.-F. Li, G. S. Samudra, C. Zhu, Z. L. Xia, X. Y. Liu, and J. F. Kang, "Strain-induced very low noise RF MOSFETs on flexible plastic substrate," in *VLSI Symp. Tech. Dig.*, 2005, pp. 160–161.
- [7] M. Kawasaki, S. Imazeki, M. Ando, Y. Sekiguchi, S. Hirota, S. Uemura, and T. Kamata, "High-resolution full-color LCD driven by OTFTs using novel passivation film," *IEEE Trans. Electron Devices*, vol. 53, no. 3, pp. 435–441, Mar. 2006.
- [8] L. Zhou, S. Park, B. Bai, J. Sun, S. C. Wu, T. N. Jackson, S. Nelson, D. Freeman, and Y. Hong, "Pentacene TFT driven AM OLED displays," *IEEE Electron Device Lett.*, vol. 26, no. 9, pp. 640–642, Sep. 2005.
- [9] K. T. Kang, M. H. Lim, H. G. Kim, Y. W. Choi, H. L. Tuller, I. D. Kima, and J. M. Hong, "Mn-doped Ba<sub>0.6</sub>Sr<sub>0.4</sub>TiO<sub>3</sub> high- $\kappa$  gate dielectrics for low voltage organic transistor on polymer substrate," *Appl. Phys. Lett.*, vol. 87, no. 24, p. 242 908, Dec. 2005.
- [10] J. H. Na, M. Kitamura, D. Lee, and Y. Arakawa, "High performance flexible pentacene thin-film transistors fabricated on titanium silicon oxide gate dielectrics," *Appl. Phys. Lett.*, vol. 90, no. 16, p. 163 514, Apr. 2007.
- [11] H. Ma, O. Acton, G. Ting, J. W. Ka, H.-L. Yip, N. Tucker, R. Schofield, and A. K.-Y. Jen, "Low-voltage organic thin-film transistors with  $\pi$ - $\sigma$ -phosphonic acid molecular dielectric monolayers," *Appl. Phys. Lett.*, vol. 92, no. 11, p. 113 303, Mar. 2008.
- [12] P. H. Wöbkenberg, J. Ball, F. B. Kooistra, J. C. Hummelen, D. M. de Leeuw, D. D. C. Bradley, and T. D. Anthopoulos, "Low-voltage organic transistors based on solution processed semiconductors and self-assembled monolayer gate dielectrics," *Appl. Phys. Lett.*, vol. 93, no. 1, p. 013 303, Jul. 2008.
- [13] M. F. Chang, P. T. Lee, S. P. McAlister, and A. Chin, "Low subthreshold swing HfLaO/pentacene organic thin-film transistors," *IEEE Electron Device Lett.*, vol. 29, no. 3, pp. 215–217, Mar. 2008.
- [14] C. H. Cheng, H. C. Pan, C. N. Hsiao, G. L. Chen, S. P. McAlister, and A. Chin, "Improved high-temperature leakage in high density MIM capacitors by using a TiLaO dielectric and an Ir electrode," *IEEE Electron Device Lett.*, vol. 28, no. 12, pp. 1095–1097, Dec. 2007.
- [15] K. C. Chiang, C. C. Huang, A. Chin, G. L. Chen, W. J. Chen, Y. H. Wu, and S. P. McAlister, "High performance SrTiO<sub>3</sub> metal-insulator-metal capacitors for analog applications," *IEEE Trans. Electron Devices*, vol. 53, no. 9, pp. 2312–2319, Sep. 2006.