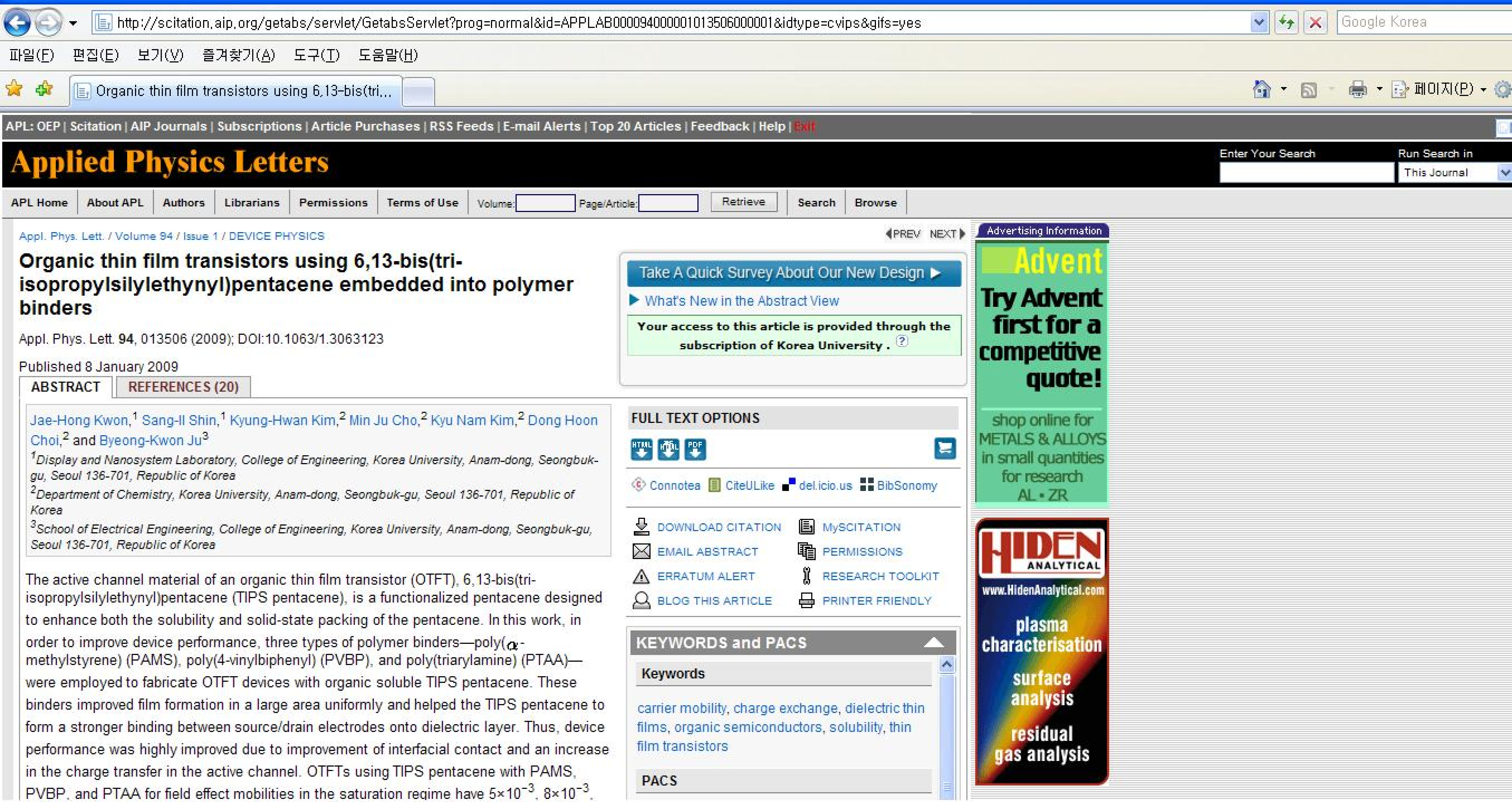

Applied Physics Letters

Appl. Phys. Lett. / Volume 94 / Issue 1 / DEVICE PHYSICS

Organic thin film transistors using 6,13-bis(tri-isopropylsilylethynyl)pentacene embedded into polymer binders

Appl. Phys. Lett. **94**, 013506 (2009); DOI:10.1063/1.3063123

Published 8 January 2009

Jae-Hong Kwon,[1] Sang-Il Shin,[1] Kyung-Hwan Kim,[2] Min Ju Cho,[2] Kyu Nam Kim,[2] Dong Hoon Choi,[2] and Byeong-Kwon Ju[3]

[1]*Display and Nanosystem Laboratory, College of Engineering, Korea University, Anam-dong, Seongbuk-gu, Seoul 136-701, Republic of Korea*

[2]*Department of Chemistry, Korea University, Anam-dong, Seongbuk-gu, Seoul 136-701, Republic of Korea*

[3]*School of Electrical Engineering, College of Engineering, Korea University, Anam-dong, Seongbuk-gu, Seoul 136-701, Republic of Korea*

The active channel material of an organic thin film transistor (OTFT), 6,13-bis(tri-isopropylsilylethynyl)pentacene (TIPS pentacene), is a functionalized pentacene designed to enhance both the solubility and solid-state packing of the pentacene. In this work, in order to improve device performance, three types of polymer binders—poly(α-methylstyrene) (PAMS), poly(4-vinylbiphenyl) (PVBP), and poly(triarylamine) (PTAA)—were employed to fabricate OTFT devices with organic soluble TIPS pentacene. These binders improved film formation in a large area uniformly and helped the TIPS pentacene to form a stronger binding between source/drain electrodes onto dielectric layer. Thus, device performance was highly improved due to improvement of interfacial contact and an increase in the charge transfer in the active channel. OTFTs using TIPS pentacene with PAMS, PVBP, and PTAA for field effect mobilities in the saturation regime have 5×10^{-3}, 8×10^{-3},

KEYWORDS and PACS

Keywords

carrier mobility, charge exchange, dielectric thin films, organic semiconductors, solubility, thin film transistors

PACS

APPLIED PHYSICS LETTERS **94**, 013506 (2009)

Organic thin film transistors using 6,13-bis(triisopropylsilylethynyl)pentacene embedded into polymer binders

Jae-Hong Kwon,[1] Sang-Il Shin,[1] Kyung-Hwan Kim,[2] Min Ju Cho,[2] Kyu Nam Kim,[2]
Dong Hoon Choi,[2] and Byeong-Kwon Ju[3,a)]
[1]*Display and Nanosystem Laboratory, College of Engineering, Korea University, Anam-dong, Seongbuk-gu, Seoul 136-701, Republic of Korea*
[2]*Department of Chemistry, Korea University, Anam-dong, Seongbuk-gu, Seoul 136-701, Republic of Korea*
[3]*School of Electrical Engineering, College of Engineering, Korea University, Anam-dong, Seongbuk-gu, Seoul 136-701, Republic of Korea*

(Received 13 August 2008; accepted 10 December 2008; published online 8 January 2009)

The active channel material of an organic thin film transistor (OTFT), 6,13-bis(triisopropylsilylethynyl)pentacene (TIPS pentacene), is a functionalized pentacene designed to enhance both the solubility and solid-state packing of the pentacene. In this work, in order to improve device performance, three types of polymer binders—poly(α-methylstyrene) (PAMS), poly(4-vinylbiphenyl) (PVBP), and poly(triarylamine) (PTAA)—were employed to fabricate OTFT devices with organic soluble TIPS pentacene. These binders improved film formation in a large area uniformly and helped the TIPS pentacene to form a stronger binding between source/drain electrodes onto dielectric layer. Thus, device performance was highly improved due to improvement of interfacial contact and an increase in the charge transfer in the active channel. OTFTs using TIPS pentacene with PAMS, PVBP, and PTAA for field effect mobilities in the saturation regime have 5×10^{-3}, 8×10^{-3}, and 2.7×10^{-2} $\mathrm{cm}^2/\mathrm{V\ s}$, respectively. © *2009 American Institute of Physics*.

[DOI: 10.1063/1.3063123]

Organic thin film transistors (OTFTs) have potential for use in many low-cost, large area, light weight, and flexible electronic applications such as integrated circuits, displays, radio-frequency identification tags, and sensor arrays. Moreover, an advantage of the OTFT is its capability of being processed from a solution at a low temperature allowing simple fabrication techniques such as spin coating, drop casting, screen printing, spray coating, and nanoimprinting.[1–5]

Pentacene has been the subject of special attention because of the combination of its commercial availability, device performance, and environmental stability. OTFTs, comprising vacuum deposited thin film pentacene providing high field effect mobility (>1 $\mathrm{cm}^2/\mathrm{V\ s}$), have been reported in many literatures.[6–9] Pentacene molecular crystals have an open herringbone structure with a combination of face to face and edge to face interactions. By attaching a functional side group, soluble and stable functionalized pentacene can be prepared,[2] thereby making standard fabrication methods more accessible for the solution processing of its organic semiconductors (OSCs) into OTFT devices.

In this work, we synthesized soluble TIPS pentacene by adding bulky groups at the 6th and 13th positions of the pentacene molecule[2] and used a 6,13-bis(triisopropylsilylethynyl)pentacene (TIPS pentacene) as an OSC for OTFT fabrication. Moreover, we prepared three kinds of polymer binder: poly(α-methylstyrene) (PAMS), poly(4-vinylbiphenyl) (PVBP), and poly(triarylamine) (PTAA). These binders mixed with the TIPS pentacene exhibit good film quality on the dielectric layer and impose function as forming a good interfacial contact between source/drain electrodes onto the active channel layer.

The device configuration of a TIPS-pentacene based OTFT is shown in Fig. 1(a). The heavy doped *p*-type Si wafer, where the 100-nm-thick dielectric was thermally grown, formed gate electrodes for the OTFTs on the Si substrates. Sequentially, gold (Au, 200 nm thickness) source/drain electrodes were deposited by electron-beam evaporation. Also, a shadow mask was used to define the source and drain electrodes. The channel width (W) and length (L) were 500 and 20 μm, respectively. Next, the TIPS-pentacene film and TIPS pentacene mixed with PAMS, PVBP, and PTAA films individually were deposited by a drop casting from a

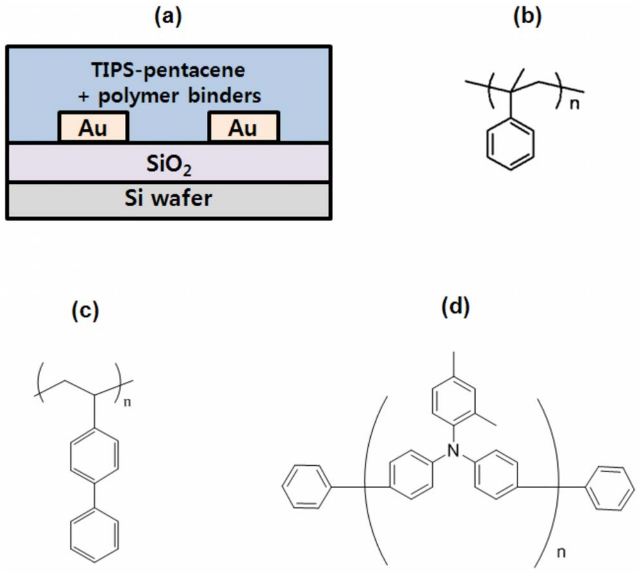

FIG. 1. (Color online) (a) A schematic of the bottom contact OTFT on a highly doped Si wafer using TIPS pentacene with polymer binders. Chemical structures of polymer binders (b) PAMS, (c) PVBP, and (d) PTAA.

[a)] Author to whom correspondence should be addressed. Electronic mail: bkju@korea.ac.kr. FAX: +82-2-3290-3791. Tel.: +82-2-3290-3237.

0003-6951/2009/94(1)/013506/3/$23.00

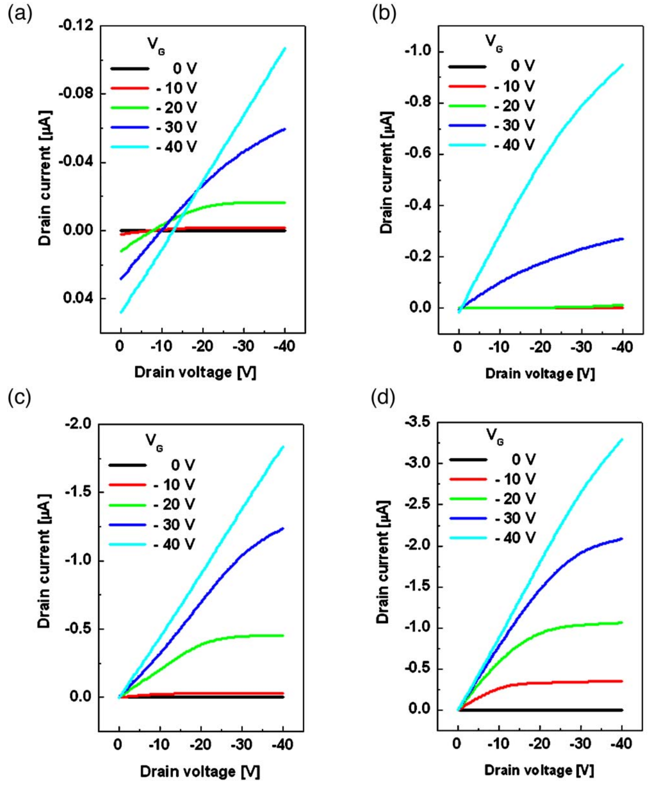

FIG. 2. (Color online) The drain current (I_D) vs the drain voltage (V_D) curves obtained from our OTFTs fabricated using TIPS pentacene (a) without a polymer binder with (b) PAMS, (c) PVBP, and (d) PTAA.

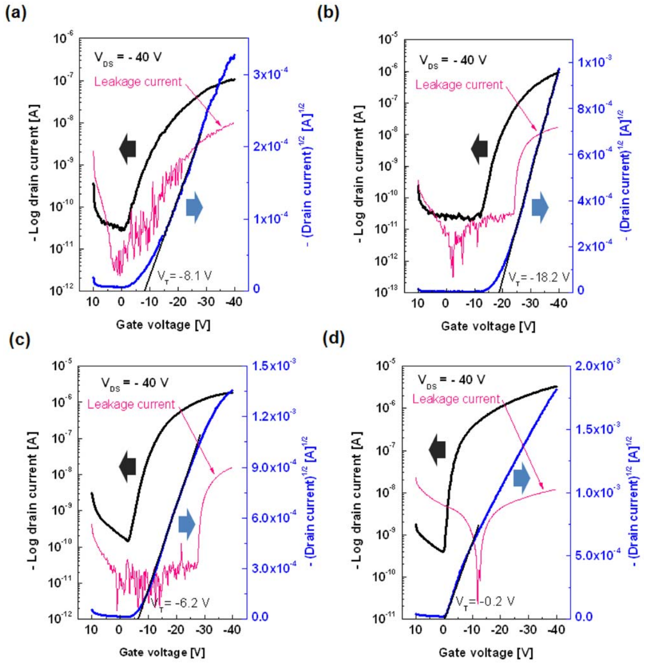

FIG. 3. (Color online) $-\sqrt{I_D}$ vs V_G and $-\log(I_D)$ vs V_G curve characteristics of the OTFTs using TIPS pentacene, (a) without polymer binder, (b) with PAMS, (c) PVBP, and (d) PTAA, that the four OTFTs obtained with $V_{DS}=-40$ V.

2 wt % solution of TIPS pentacene in monochlorobenzene, respectively. Finally, after coating the OSC, the device was annealed using a hotplate at 110 °C for 1 min. After the device fabrication, we measured the transistor characteristics using a Keithley SCS/4200 in a dark box.

Figures 1(b)–1(d) show the chemical structure of PAMS (Sigma-Aldrich, used as received), PVBP (Sigma-Aldrich, used as received), and PTAA (used as synthesized), respectively. Moriguchi *et al.*[10] reported that the crystal quality was improved by using a PAMS treatment so that the carrier mobility was higher than that of nontreatment since the chain dynamics of the PAMS of high molecular weight in benzene, a good solvent, in the dilute solution served as the hydrodynamic interaction of flexible linear polymers in solution.[11] In addition, Brown *et al.*[12] proposed that the TIPS pentacene comprised polymer binders of PAMS or PVBP. In other words, these preferred insulator binders act as an adhesion layer as well as a nonpolar capping layer to overcome the problem of poor compatibility between the TIPS pentacene and the gate dielectric layer at their interface.[13] Schroeder *et al.*[14] reported a method of decreasing the injection barrier from the metal electrode to the OSC, namely, pentacene and PTAA as the OSC. Here, PTAA acts as a good hole conductor between the OSC and source and drain electrodes as reducing surface dipoles.[15]

Figures 2(a)–2(d) show the drain current (I_D)–drain voltage (V_D) curves of our OTFTs using TIPS pentacene without a polymer binder and with PAMS, PVBP, and PTAA, respectively. These curves are typical *p*-type OTFTs working in an accumulation mode. According to the figures, the highest drain current of −3.3 μA was obtained from the OTFT using TIPS pentacene mixed with PTAA with a gate bias (V_G) of −40 V while the OTFT using TIPS pentacene mixed with PVBP and PAMS, and without a polymer binder showed the highest drain current of −1.84, −0.94, and −0.11 μA, respectively (at $V_G=-40$ V). Figure 2(a) shows the I_D-V_D characteristics of our standard TIPS-pentacene small molecule without a polymer binder. The drop casted TIPS-pentacene film was approximately 300 nm thick and was not patterned for these devices. The presence of large drain current offset[16] in a low gate bias is due to leakage current in the output curve because of poor film forming property in TIPS pentacene only. The poor interfacial contact between active layer and dielectric layer can be expected. Figure 2(b) shows the I_D-V_D characteristics of the OTFT using TIPS pentacene mixed with PAMS. As expected, for the transistor, the output characteristics showed two distinct regions of device operation: linear and saturation. However, Fig. 2(b) shows the relatively nonideal comparison behavior of the device with the linear and saturation regime in Figs. 2(c) and 2(d). For that reason, we think that the minor current-crowding effect might be due to the contact resistance between the OSC channel and source and drain electrodes.[17] Furthermore, the threshold voltage ($V_T=-18.2$ V) of the TIPS-pentacene based OTFT with PAMS appeared higher than other devices with polymer binders such as PVBP and PTAA since PAMS offered improving solubility and continuity of similar dielectric properties.[18] Also, the I_D-V_D characteristics for both (c) and (d) in Fig. 2 exhibited pinch-off and current saturation.

Figures 3(a)–3(d) show transfer characteristics of our OTFTs using TIPS pentacene without a polymer binder and with PAMS, PVBP, and PTAA, respectively. The field effect mobility was calculated from these data in the saturation regime ($V_{DS}=-40$ V) by plotting the $-\sqrt{I_D}$ versus V_G and fitting the data to the following equation:[19]

TABLE I. The OTFT characteristics of the field effect motilities in the saturation regime, the threshold voltages, on/off current ratios, and the subthreshold swings for various polymer binders.

	μ (cm^2/V s)	V_T (V)	$I_{\text{on/off}}$	SS (V/decade)
Without polymer binder	0.001	−8.1	4.33×10^3	2.38
PAMS	0.005	−18.2	5.51×10^4	1.19
PVBP	0.008	−6.2	1.25×10^4	2.52
PTAA	0.027	−0.2	8.42×10^3	0.36

$$I_{DS}=(WCi/2L)\mu(V_G-V_T)^2,$$

where $C_i=3.45\times10^{-8}$ F cm^{-2}, $W=500$ μm, and $L=20$ μm. The V_T can also be determined in the OTFT saturation regime by plotting $-\sqrt{I_D}$ versus V_G and extrapolating the tangential line drawing, namely, intercept on the x-axis, illustrated in Figs. 3(a)–3(d). Additionally, on/off current ratio ($I_{\text{on/off}}$) and subthreshold swing (SS) were determined from $-\log I_D$ versus V_G curves of Figs. 3(a)–3(d). Such parameters for all our devices are summarized in Table I.

The device with PTAA showed the best overall performance except for the $I_{\text{on/off}}$ and I_{off}. The highest mobility (0.027 cm^2/V s), the lowest V_T, and the smallest SS value were achieved for the sample with PTAA. For the $I_{\text{on/off}}$ of the sample with PAMS, we found that this sample was much better than other samples with polymer binders and the I_{off} was lower than other samples with polymer binders. It should be noted that although the OTFT with PTAA exhibited the highest mobility, it suffered from a low $I_{\text{on/off}}$. This low ratio arose from the high I_{off} indicating a high leakage current possibly due to a high trap density at the interface between active layer and dielectric layer.[20]

In summary, we fabricated four different OTFTs using TIPS pentacene with polymer binders such as PAMS, PVBP, and PTAA that were fabricated on a SiO$_2$ dielectric/Si wafer substrate. First, the device with PAMS presented a more stable performance than the reference device. However, this device appeared to have larger V_T than other devices due to the intrinsic low charge mobility of the OSC. That is, the field-induced current was proportional to the field-induced charge density. It was likely that this device needed a greater surface charge density at the OSC/dielectric interface. Second, the SS of the device with PVBP showed lower properties than other devices except for the reference device. A large SS generally implied a large concentration of shallow traps. That is, this device needed to improve properties of interface quality and to have the high density of trap states on the OSC/dielectric interface. Finally, the $I_{\text{on/off}}$ of the device with PTAA was lower than other devices with polymer binders, indicating a high leakage. To block the leakage current, this device can have a reduced trap density without adding any more trap sites in OSC/dielectric interface using Ar ion beam treatment.[20]

This work was supported by the National Research Laboratory (NRL) (Contract No. R0A-2007-000-20111-0), Program of the Ministry of Science and Technology (Korea Science and Engineering Foundation), the Korea Science and Engineering Foundation (KOSEF) grant funded by the Korea Ministry of Education, Science and Technology (MEST) (Grant No. R11-2007-045-01003-0), and the second stage of the Brain Korea 21 Project in 2008 (Korea Research Foundation).

[1] J. H. Kwon, J. H. Seo, H. Kang, D. H. Choi, and B. K. Ju, J. Appl. Phys. **101**, 064502 (2007).

[2] J. H. Kwon, J. H. Seo, S. I. Shin, K. H. Kim, D. H. Choi, I. B. Kang, H. Kang, and B. K. Ju, IEEE Trans. Electron Devices **55**, 500 (2008).

[3] J. H. Kwon, S. H. Lee, and B. K. Ju, J. Appl. Phys. **101**, 104515 (2007).

[4] J. H. Kwon, K. S. Lee, Y. H. Lee, and B. K. Ju, Electrochem. Solid-State Lett. **9**, H85 (2006).

[5] M. D. Austin and S. Y. Chou, Appl. Phys. Lett. **81**, 4431 (2002).

[6] H. Klauk, M. Halik, U. Zschieschang, G. Schmid, W. Radlik, and W. Weber, J. Appl. Phys. **92**, 5259 (2002).

[7] Y. Kato, S. Iba, R. Teramoto, T. Sekitani, T. Someya, H. Kawaguchi, and T. Sakurai, Appl. Phys. Lett. **84**, 3789 (2004).

[8] G. Wang, Y. Luo, and P. H. Beton, Appl. Phys. Lett. **83**, 3108 (2003).

[9] B. Stadlober, M. Zirkl, M. Beutl, G. Leising, S. B. Gogonea, and S. Bauer, Appl. Phys. Lett. **86**, 242902 (2005).

[10] N. Moriguchi, T. Nishikawa, T. Anezaki, A. Unno, M. Tachibana, and K. Kojima, Physica B **376**, 276 (2006).

[11] Y. Tsunashima, J. Chem. Phys. **110**, 12211 (1999).

[12] B. A. Brown, J. Veres, and S. D. Ogier, U.S. Patent No. 0,259,477 (4 October 2007).

[13] C. Yang, K. Shin, S. Y. Yang, H. Jeon, D. Choi, D. S. Chung, and C. E. Park, Appl. Phys. Lett. **89**, 153508 (2006).

[14] R. Schroeder, L. A. Majewski, M. Grell, J. Maunoury, J. Gautrot, P. Hodge, and M. Turner, Appl. Phys. Lett. **87**, 113501 (2005).

[15] N. Koch, J. Ghijsen, A. Elschner, R. L. Johnson, J.-J. Pireaux, J. Schwartz, and A. Kahn, Appl. Phys. Lett. **82**, 70 (2003).

[16] H. Jia, G. K. Pant, E. K. Gross, R. M. Wallace, and B. E. Gnade, Org. Electron. **7**, 16 (2006).

[17] R. R. Troutman and A. Kotwal, IEEE Trans. Electron Devices **36**, 2915 (1989).

[18] G. Erwann, B. Peter, F. Walter, R. Henning, and U. Andreas, U.S. Patent No. 7,298,023 (5 September 2007).

[19] P. Servati, A. Nathan, and G. A. J. Amaratunga, Phys. Rev. B **74**, 245210 (2006).

[20] S. Kang, S. Jung, J. Park, H. J. Lee, and M. Yi, Microelectron. Eng. **84**, 1503 (2007).