Flexible Low-Power Operative Organic Source-Gated Transistors

Yonghee Kim, Eun Kwang Lee, and Joon Hak Oh*

Low-voltage operation and fast switching ability are necessary for wearable electronic devices. Recently, electrolyte dielectric materials have been widely used to decrease driving voltages; however, they often exhibit unwanted doping effects and power dissipation problems. Here, a method for dramatically lowering driving voltages is reported in organic electronics via source-gated transistor (SGT) structures. SGTs are fabricated by evaporating asymmetric metals with different work functions for the source and drain electrodes. Versatile organic semiconductor-based SGTs demonstrate a significantly lower drain voltage (<10 V) for the saturation regime compared to that of typical field-effect transistors with the same dielectric layer (>80 V). Furthermore, coating reduced Pyronin B (rPyB) onto n-type SGTs decreases the threshold voltage from 51.2 to 0.1 eV and improves air-stability, exhibiting a maintained electron mobility (>90%) for 40 d. The air-stability is due to both the energetic and kinetic factors, including a decreased lowest unoccupied molecular orbital level of the n-type semiconductor after doping and covering the active layer with rPyB. Finally, flexible SGTs are fabricated on a Parylene-C substrate that shows highly stable operation in a bending test. The results demonstrate a promising technology for low-power, flexible electronic devices via electrode engineering.

1. Introduction

In recent years, there has been an increasing need for wearable devices, which are a key platform in the Internet of Things (IoT) technologies. Organic semiconducting materials (π-conjugated molecules) have been recognized as one of the key active-layer materials for wearable devices,[1–3] due to their intrinsic mechanical flexibility, light weight, easily controllable molecular structures, and low processing cost. To date, many studies on organic field-effect transistors (OFETs) have focused on the development of new organic semiconductors (OSCs),[4–6] leading to significant improvements in device performance. The best electrical performance for OFETs has now surpassed a charge carrier mobility of over 10 cm² V⁻¹ s⁻¹.[7–10]

However, such wearable devices require both excellent electrical performance and low-power operation in practical applications. In particular, low-power operation is important because there is a limit to the supplied energy in wearable devices. To date, however, most OFETs have a high threshold voltage ($V_{th}$) and a high saturation drain voltage ($V_{DS,sat}$), which limit the commercialization of OFET-based wearable devices.

To resolve these problems, a number of studies have investigated the development of dielectric materials in field-effect transistors (FETs). First, high-permittivity (high-$k$) dielectric materials such as inorganic metal oxide materials such as $\text{TiO}_2, \text{Al}_2\text{O}_3, \text{HfO}_2$ have been developed for FETs. However, these materials have poor mechanical characteristics for use in soft electronics.[14] Furthermore, a thin dielectric layer often leads to undesirable gate leakage current. Second, electrolyte materials, such as a salt,[15] an ionic liquid,[16] an ion gel,[17] and a copolymer,[18] have been used for the gate dielectric. This allows very low voltage operation; however, there are some limitations, such as delamination of the materials in contact with such dielectrics, unwanted doping effects induced by ion penetration, and power dissipation problems.[19,20]

As an alternative to dielectric materials, an approach based on simple modification of the device structure has been studied. In source-gated transistors (SGTs), asymmetric electrodes with different work functions (WFs) were used for the source and drain electrodes. SGTs with low $|V_{DS,sat}|$ have been fabricated using inorganic materials, such as amorphous silicon,[21] polysilicon,[22] and zinc oxide.[23,24] In SGTs, the Schottky barrier between the source electrode and the semiconductor controls charge carrier transport in the active layer. Due to this barrier, only charge carriers with a high electric field can move from the source electrode to the drain electrode. This effect leads to operational advantages such as significantly decreased $|V_{DS,sat}|$ and a flat saturation drain current ($I_{DS}$).[22,25,26] Moreover, Silva and co-workers reported an SGT-based inverter circuit with high intrinsic gain ($G = g_m/g_d$, where the transconductance...
\[ g_m = \frac{dI_{DS}}{dV_{GS}}, \] the gate voltage is \( V_{GS} \), and the output conductance \( g_0 = \frac{dI_{DS}}{dV_{DS}} \) because SGTs have very low \( g_0 \) due to the flat saturated \( I_{DS} \) based on simulation results.\(^{[27]}\)

However, the Schottky barrier can decrease the charge density in the active layer. In particular, \( V_{GS} \) is related to filling the trap energy states in the channel\(^{[28]}\) and tends to be high in SGTs. With a goal to reduce \( V_{GS} \), Silva et al. introduced doping of the inorganic semiconductor by annealing dopants at 550 °C.\(^{[27]}\) However, such high temperature damages the lattice structure of OSCs, leading to reduced charge carrier mobility.\(^{[29]}\) A low-temperature process is required for fabricating low-power flexible electronic devices on plastic substrates. Therefore, the inorganic semiconductor-based SGTs developed thus far are not highly suitable for use in flexible low-power electronics, and it is necessary to develop SGTs based on OSCs. Although Guo and Shannon reported a pentacene-based SGT using simulation methods,\(^{[30]}\) there has been no report on the experimental demonstration and analysis of organic source-gated transistors (OSGTs).

Here, we report the first experimental demonstration of flexible, low-voltage operative SGT by synergistically combining an OSC-based SGT with a molecular doping method. We fabricated OSGTs based on four representative small molecular and polymeric semiconductors (pentacene, BPE-PTCDI, PDBT-co-TT, and N2200) at a low temperature (<150 °C). All the OSGTs exhibited a remarkably lower \( |V_{DS, sat}| \) in comparison with the classic OTFETs based on the same semiconductor and dielectric layers, revealing high effectiveness and universal applicability of the developed device platform for flexible low-power electronics. Notably, the developed n-type molecular doping greatly reduced the \( |V_{GS}| \) of the OSGTs based on n-type polymeric semiconductor N2200 and maintained the electron mobility of the devices up to 93.2% for 40 d under ambient conditions. This doping effect was studied by measuring the lowest unoccupied molecular orbital (LUMO) level of the n-type doped semiconductors using ultraviolet photoelectron spectroscopy (UPS) and ultraviolet-visible (UV-vis) spectroscopy. Finally, flexible 7 \( \times \) 7 OSGT arrays on a Parylene-C substrate were fabricated, which maintained the electron mobility up to 94.6% under a bending radius of 5.8 mm. Our results demonstrate an efficient way to develop low-power electronic devices based on organic materials that can expedite the commercialization of deformable organic electronic devices.

### 2. Results and Discussion

#### 2.1. Working Principles of OSGTs

In this work, four kinds of OSCs with different derivatives were used for the OSGTs. Pentacene (polycyclic aromatic hydrocarbon, p-type) and N,N′-bis(2-phenylethyl)-perylene-3,4,9,10-tetracarboxylic diimide, BPE-PTCDI (PTCDI derivative, n-type) were used for small molecular OSCs. Poly\{2,2′-(2,5-bis(2-octyldodecyl)-3,6-dioxo-2,3,5,6-tetrahydropyrrolo[3,4-c]pyrrole-1,4-diyl) dithiophene-5,5′-diyl-alt-thieno[3,2-b] thiophen-2,5-diyl\}, DPP-based polymer, ambipolar), and poly\{N,N′-bis(2-octyldodecyl)-naphthalene-1,4,5,8-bis(dicarboximide)-2,6-diyl-alt-5,5′-(2,2′-bithiophene)\}, P\{NDI2OD-T2\}, N2200 (NDI-based polymer, n-type) were used for the polymeric OSCs. Before fabricating OSGTs using these OSCs, molecular orbital energy level analyses were conducted. The energy level results (Figure 1a; Figure S1a–d, Supporting Information) were calculated by UPS (Figure S2a–d, Supporting Information) that measured vacuum level shifts from Au and the highest occupied molecular orbital (HOMO) energy level for the materials at a base pressure of 10−10 Torr using He I excitation lines (\( h\nu = 21.22 \) eV) as well as by UV-vis spectroscopy (Figure S3, Supporting Information) that measured the bandgap of materials for information on the LUMO energy level. From these results, we chose three electrodes for use in the OSGTs: aluminum (Al) with a WF of 4.15 eV (Figures S1e and S2e, Supporting Information) close to the LUMO level of N2200 (−4.09 eV), BPE-PTCDI (−3.97 eV), and PDBT-co-TT (−4.35 eV) as the drain electrode in n-type OSGTs; gold (Au) with a WF (5.10 eV) close to the HOMO level of pentacene (−5.15 eV) as the drain electrode in pentacene-based p-type OSGTs; and selenium (Se) with a WF (5.90 eV) close to the HOMO level of PDBT-co-TT (−5.75 eV) for PDBT-co-TT based p-type OSGTs. Density functional theory (DFT) calculation results using the Gaussian09 software showed similar trends to the experimental values in the energy levels of these OSCs (Figure S4, Supporting Information).

OSGTs were fabricated using asymmetric source and drain electrodes in a bottom-gate top-contact configuration, as shown in Figure 1b–d. First, OSC was deposited on a n-octadecyltrimethoxysilane (OTS)-treated SiO2/Si wafer (300 nm-thick SiO2) using spin-coating for the polymeric semiconductors or thermal evaporation under a high vacuum (<5 × 10−6 Torr) for the small molecular semiconductors. Next, Al for the drain electrode and Au (or Se) for the source electrode were deposited separately on n-type semiconductors using thermal evaporation, whereas Al for the source electrode and Au (or Se) for the drain electrode were deposited separately on p-type semiconductors. The detailed fabrication procedure is described in the Experimental Section. We studied the mechanism of operation of OSGTs (Figure 1e). In N2200 OSGTs, Al acted as an Ohmic contact and Au acted as a Schottky contact with regard to the LUMO level of N2200 (Figure 1f). As shown in Figure 1g, the current \( (I_{DS}) \) and voltage \( (V_{DS}) \) exhibited a linear relationship, confirming the Ohmic contact when using Al electrodes for both source and drain electrodes in devices (blue line). In contrast, rectifying contact characteristics were shown for Au electrodes (red line). In the region between the source electrode and N2200, a Schottky barrier was generated because of the high gap between the LUMO of N2200 and the WF of gold. Due to this barrier, the charge carriers (electrons) in n-type semiconductor devices must overcome or penetrate this barrier to reach the drain electrodes, and only charge carriers under a high electric field can move through the barrier, so OSGTs exhibited a lower \( V_{DS, sat} \) for the saturation regime than OTFETs.

To study the effective Schottky barrier between the semiconductor and source electrode according to \( V_{GS} \) for OSGT operation, \( I_{DS}-V_{DS} \) was measured in the temperature range of 25–105 °C at \( V_{GS} = 0 \) and 10 V (Figure 2a,c). When the temperature increased, the Schottky barrier height \( (H_{SB}) \) decreased, leading to increased \( I_{DS} \). The effective \( H_{SB} \) for a value of \( V_{GS} \)
of 0 V (diode mode) was calculated using the thermionic emission model\textsuperscript{[23,31]} as

\[ I_{DS} = \frac{RAT^2e^{\frac{qV_{DS}}{kT}}}{1 + \frac{qV_{DS}}{kT}} \]  

(1)

where \( R = 1.202 \times 10^{-6} \, \text{A m}^{-2} \, \text{K}^{-2} \) is the Richardson constant, \( A \, [\text{m}^2] \) is the effective contact area, \( T \, [\text{K}] \) is the absolute temperature, \( q = 1.602 \times 10^{-19} \, \text{C} \) is the elementary electronic charge, and \( k_B = 8.617 \times 10^{-5} \, \text{eV K}^{-1} \) is Boltzmann's constant. From Equation (1), \( H_{SB} \) was calculated from the slope of \( \ln(I_{DS}/T^2) \) on \( 1/(k_B T) \) (Arrhenius plot) and found to be 0.2407 eV (Figure 2b). When a value of \( V_{GS} \) of 10 V was applied to the OSGTs (transistor mode), \( H_{SB} \), which is the same as the activation energy, was calculated using the Arrhenius equation

\[ I_{DS} = Ce^{\frac{qV_{DS}}{k_B T}} \]  

(2)

where \( C \) is a constant factor.\textsuperscript{[26]} From Equation (2), \( H_{SB} \) was calculated from the slope of \( \ln(I_{DS}) \) versus \( 1/(k_B T) \); the value

![Figure 1. a) Energy alignment of OSC materials and metals. Fabrication of OSGTs (from insets (b)–(d)): b) OSC was deposited on the OTS-treated oxidized Si wafer (300 nm SiO\textsubscript{2}). c,d) Source and drain electrodes were deposited through a shadow mask on the semiconductor film using a thermal evaporator. e) Schematic of the device structure of N2200 OSGTs and surface charge transfer doping using spin-coated rPyB. f) Energy diagram of N2200 OSGTs. g) \( I_{DS}-V_{DS} \) characterization for Ohmic contact and Schottky contact.](image-url)
was 0.1681 eV (Figure 2d). From these results, $H_{SB}$ in the OSGTs (Figure 2e) was changed according to the electric field between the gate electrode and the source electrode. If no gate voltage was applied, charge carriers moved through the relatively higher Schottky barrier height ($H_{SB,1}$) under the thermionic emission model. However, when $V_{GS}$ was applied, charge carriers passed through the lower Schottky barrier height ($H_{SB,2}$) under the thermionic-field emission model (Figure 2f). These results showed that OSCs can be applied to OSGTs.

2.2. Electrical Performances of OSGTs Based on Various OSCs

To demonstrate the electrical properties of OSGTs, the transfer curves ($I_{DS}$–$V_{GS}$) and output curves ($I_{DS}$–$V_{DS}$) of the OFETs and OSGTs were compared. The electrical characteristics of OFETs and OSGTs were measured under an N$_2$ environment. The transistors were driven in the saturation regime ($V_{DS} = \pm 100$ V for OFETs, $V_{DS} = \pm 10$ V or $\pm 20$ V for OSGTs, and the charge carrier mobility, $\mu$, was calculated in the saturation regime as

$$\mu = \frac{2L}{W} \frac{I_{DS}}{C_{ox} (V_{GS} - V_{th})}$$

where $L$ is the channel length, $W$ is the channel width, and $C_{ox}$ is the capacitance per unit area of the total gate dielectric layer. The typical transfer and output curves were measured under the following parameters: $C_{ox} = 1.15 \times 10^{-8}$ F cm$^{-2}$ for 300 nm SiO$_2$, $W = 180$ and $L = 90$ $\mu$m for OFETs; $W = 440$ and $L = 110$ $\mu$m for OSGTs.

We fabricated OSGTs using pentacene (p-type small molecule), BPE-PTCDI (n-type small molecule), PDBT-co-TT (amphipolar polymer), and N2200 (n-type polymer). First, OSGTs based on pentacene and BPE-PTCDI were fabricated using a thermal evaporator as described in the Experimental Section. These OSGTs showed a decrease in the $V_{DS,sat}$ ($< 20$ V) (Figure 3)
compared to OFETs (>80 V) (Figure S5, Supporting Information) caused by the Schottky barrier, as described in Section 2.3. (vide infra). Both \( I_{\text{on}} \) and \( I_{\text{off}} \) were reduced, while \( I_{\text{on}} \) significantly decreased. Due to the decreased \( I_{\text{on}} \), the hole or electron mobility was decreased compared to conventional OFETs. Furthermore, OSGTs based on PDBT-co-TT showed high \( I_{\text{on}}/I_{\text{off}} \) and low \( |V_{\text{DS,sat}}| \) (<20 V) due to the Schottky barrier (Figure 4) compared to that of typical OFETs (<100 V) (Figure S6, Supporting Information). In the p-type (n-type) operation, Se was used as the drain (source), and Al was used as the source (drain) electrode. The ambipolar OSGTs showed greatly improved \( I_{\text{on}}/I_{\text{off}} \) (10^5–10^7) (Table 1). In the ambipolar active layer, the holes moved from Al to Se and the electrons moved from Se to Al. Because the Schottky barrier that can lower the \( I_{\text{off}} \) was generated in the source electrode region, the \( I_{\text{on}}/I_{\text{off}} \) can be increased. To avoid the overestimation issues of charge carrier mobility, we calculated the measurement reliability factor \( r \) in the saturation regime, as shown in Figures 3 and 4, Table 2, and Figures S5 and S6 (Supporting Information). The parameter \( r \) [%] is defined as the ratio of the maximum FET conductivity achieved in experiments (\( \mu_{\text{measured}} \)) to the maximum ideal FET conductivity (\( \mu_{\text{ideal}} \))

\[
\begin{align*}
    r & = \frac{\mu_{\text{measured}}}{\mu_{\text{ideal}}} = \left( \frac{I_{\text{DS,max}}^{0.5} - I_{\text{DS,0}}^{0.5}}{V_{\text{G,max}} - V_{\text{G,0}}} \right)^2 \left( \frac{W/C_{\text{ox}} \mu_{\text{ideal}}}{2L} \right) 
\end{align*}
\]

where \( I_{\text{DS,max}} \) is the experimentally measured maximum source–drain current reached at the maximum gate voltage, \( V_{\text{G,max}} \). The parameter \( I_{\text{DS,0}} \) indicates the source–drain current at the amplification onset gate voltage, \( V_{\text{G,0}} \). The nonlinearity of the OSGTs in the \( I_{\text{DS}}^{0.5}–V_{\text{GS}} \) results may be due to the Schottky barrier between OSCs and electrodes that controls the charge carriers in the active channel of OSGTs.

### 2.3. OSGTs Based on n-Type Polymer with Low \( V_{\text{th}} \) and High Air-Stability

As described in Section 2.2., the OSGTs exhibited a lower \( |V_{\text{DS,sat}}| \) and an increased \( |V_{\text{G}}| \) compared with those of a typical OFET. However, both \( V_{\text{DS}} \) and \( V_{\text{G}} \) for operation should be at a low voltage level to realize low-power application. To decrease \( V_{\text{th}} \) in n-type transistors, rPyB (Figure 1e) was used as the n-type dopant. The rPyB solution in hexane was synthesized as described in previous work. The rPyB solutions were made at concentrations of 0.1, 0.5, 0.7, and 1.0 mg mL\(^{-1}\). The solutions were spin-coated on the surface of N2200 OSGTs to give hydride-mediated n-type doping effect.

To demonstrate the electrical properties of n-type doped OSGTs, the transfer curves (\( I_{\text{DS}}–V_{\text{GS}} \)) and output curves

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Figure 3. Electrical characteristics of OSGTs based on small molecules. a) Transfer curve of pentacene OSGTs (inset: OSGT structure). b) Output curves of pentacene OSGTs (inset: \( I_{\text{DS}}–V_{\text{DS}} \) curve that ranged from 0 to –20 V of \( V_{\text{DS}} \)). c) Transfer curve of BPE-PTCDI OSGTs (inset: OSGT structure). d) Output curves of BPE-PTCDI OSGTs (inset: \( I_{\text{DS}}–V_{\text{DS}} \) curve ranging from 0 to 20 V of \( V_{\text{DS}} \)).
(\(I_{DS-V_{DS}}\)) of the rPyB-doped N2200 OFETs and OSGTs were compared (Figure 5a–d and Table 3). The transistors were operated in the saturation regime (\(V_{DS} = 100\) V for OFETs, \(V_{DS} = 10\) V for OSGTs). The transfer curve of N2200 OSGTs (Figure 5a) in the top contact bottom gate (TCBG) structure showed a dramatic change in \(V_{th}\) and an increase in current as the dopant concentration increased. In particular, 0.7 mg mL\(^{-1}\) rPyB-doped OSGTs showed very low \(V_{th}\) (0.1 V). Furthermore, \(V_{DS,sat}\) in OSGTs was very low (<10 V) (Figure 5b). For comparison, the transfer curve of N2200 typical OFETs (Figure 5c) in the same structure showed a highly reduced \(I_{on}/I_{off}\) due to the increased off current as the concentration of the n-type dopant increased. The value of \(V_{DS,sat}\) in OFETs was significantly higher (>80 V) compared to OSGTs (Figure 5d). These results indicate that low-voltage operation and fast saturation were possible in the OSGTs due to the Schottky barrier. From these OFET and OSGT results, we calculated the saturation power density (\(P_{sat}\), \(I_{DS} \times V_{DS}/(W \times L)\)) in the saturation regime according to the dopant concentration in OFETs and OSGTs (Figure 5e). The \(P_{sat}\) in OSGTs was lower by two orders of magnitude compared to that in OFETs because a low \(V_{DS}\) was needed to induce the saturation regime. Furthermore, a very low \(V_{GS}\) was required for saturation operation in OSGTs because \(V_{th}\) was very low after doping (Figure 5f, inset). Since \(V_{th}\) is the required gate voltage to fill the trap density in OSC, this result indicates that the n-type doping effect was effective.

Table 1. Electrical characteristics of OSGTs based on PDBT-co-TT compared to those of typical OFETs. The p-channel and n-channel characteristics of OFETs were measured at \(V_{DS} = -100\) and 100 V, respectively. The p-channel and n-channel characteristics of OSGTs were measured at \(V_{DS} = -20\) and 20 V, respectively.

<table>
<thead>
<tr>
<th>Device structure</th>
<th>(I_{on}/I_{off})(^a)</th>
<th>(V_{th, h}) [V](^b)</th>
<th>(\mu_h) [cm(^2) V(^{-1}) s(^{-1})](^c)</th>
<th>(I_{on}/I_{off})(^d)</th>
<th>(V_{th, e}) [V](^e)</th>
<th>(\mu_e) [cm(^2) V(^{-1}) s(^{-1})](^f)</th>
</tr>
</thead>
<tbody>
<tr>
<td>OFET</td>
<td>(10^3 \text{ to } 10^4)</td>
<td>-8.8</td>
<td>1.19 (± 0.23)(^i)</td>
<td>(10^2 \text{ to } 10^3)</td>
<td>57.8</td>
<td>2.43 (± 0.11)(^i)</td>
</tr>
<tr>
<td>OSGT</td>
<td>(10^4 \text{ to } 10^2)</td>
<td>-7.9</td>
<td>2.25 (± 0.02) x 10(^{-1})</td>
<td>(10^2 \text{ to } 10^3)</td>
<td>44.5</td>
<td>8.65 (± 0.39) x 10(^{-2})</td>
</tr>
</tbody>
</table>

\(^a\)On/off current ratio of p-channel; \(^b\)Threshold voltage of p-channel; \(^c\)Charge carrier mobility of p-channel; \(^d\)On/off current ratio of n-channel; \(^e\)Threshold voltage of n-channel; \(^f\)Charge carrier mobility of n-channel; \(^i\)The standard deviation.
and useful for OSGTs. The filled trap site density ($\Delta N_s$) arising from n-type doping was calculated from the change in the value of $V_{th}$ using

$$\Delta N_s = \frac{C_{ox} (V_{th} - V_{th,doped})}{q}$$  \hspace{2cm} (5)$$

This high-filled trap-site effect in OSGTs (Figure 5f) may occur because of the Schottky barrier in the source electrode region, which causes deficient charge carriers in the active layer. From n-type doping, charge carriers were supplemented in the active layer to fill the trap sites, and $V_{th}$ was dramatically changed according to the n-type dopant concentration. However, the output characteristics of OSGTs showed that n-type doping did not affect $V_{DS,sat}$ to reach a saturated $I_{DS}$ (Figure S7, Supporting Information). In addition, the output characteristics of the doped OSGTs showed a flat saturation current effect caused by the Schottky barrier, which provides a nearly constant number of charge carriers from the source electrode, similar to the pristine OSGTs.

The air-stability of n-type semiconductor-based devices is an important property for their commercialization because n-type transistors are typically unstable in air. Empirically, as the LUMO level of n-type semiconductors is higher than $4.0 \text{ eV}$, electrons in the semiconductor can be trapped in water and oxygen in air. [35,36] To analyze the air-stability, the devices were left in an ambient environment for several days, and the electrical characteristics were measured in air. The electron mobilities of rPyB-coated N2200 OSGTs were maintained up to 93.2% for 40 d under ambient conditions (Figure 6a,b). There are two reasons why the rPyB coating increased the stability of the electron mobility under ambient conditions. First, the dopant can supply charge carriers (electrons) to n-type semiconductors using a hydride-mediated surface doping process. To investigate the energy level alignment of rPyB-doped N2200, UPS, and UV–vis spectroscopy were performed (Figure 6c–e; Figure S8, Supporting Information). Detailed results of the UPS and UV–vis spectra are provided in Figures S9 and S10 in the Supporting Information. The high binding energy cutoff was used to calculate the vacuum level shift $\Delta E$ and the shift in the binding energy relative to the Fermi level of the gold layer was used to calculate HOMO levels in N2200. [37] The optical bandgap of the N2200 film was measured using the UV–vis absorption spectra. The measured bandgap values of N2200 were very similar, regardless of dopant concentration. Using these bandgap values, the LUMO levels of N2200 were determined. [38] The broad value of $\Delta E$ with increasing n-type dopant concentration indicated an increasing n-type doping effect. [39] Based on these results, the LUMO level decreased with n-type doping, making the n-type semiconductor devices air-stable. Second, the dopant layer covered the N2200 active channel; the thickness of the rPyB layer measured using atomic force microscopy (AFM) was $4.4 \text{ nm}$ (Figure S11, Supporting Information), which might act as a kinetic barrier to decrease the diffusion of ambient oxidants into the channel area of the N2200 layer. The surface n-type doping method developed herein did not damage the underlying polymer semiconducting layer because of the solvent orthogonality (n-hexane) (Figure S12, Supporting Information).

In addition, we also compared three types of doping processes on N2200 FETs: (i) bulk doping (rPyB was blended with N2200), (ii) contact doping (rPyB was coated on the space between the electrode and the semiconductor), and (iii) surface doping (rPyB was coated only on the active layer) (Figure S13 and Table S1, Supporting Information). The concentration of the rPyB solution used for comparison was 0.1 mg mL$^{-1}$. The transfer curves of the three types of doping processes were compared. Although the strongest doping effect was observed from the bulk doping, the $I_{on}/I_{off}$ was substantially reduced with increased $I_{on}$. Similarly, a high doping effect was observed from the surface doping, together with the remaining high $I_{on}/I_{off}$. Furthermore, we measured the contact resistance ($R_c$) using the transfer line method under different values of $L$ (Figure S14, Supporting Information) using $R_{total} = R_c \times W + R_{channel} \times L$, where $R_{total}$ is the total resistance and $R_{channel}$ is the resistance in the channel (active layer). At the point of zero $L$ in the line plot ranging from 50 to 350 $\mu$m, the surface doping yielded the smallest $R_c \times W$ value (3.09 $\times 10^4$ $\Omega$ cm). On the other hand, the contact doping produced an increased $R_c \times W$ value, which may be because positively charged PyB, which is an oxidized form of rPyB after donating electrons to the active layer, hindered the contact area of the electrode during channel doping and acted as a charge scattering site or insulating layer. In addition, bulk doping may affect the crystallinity of N2200 when dopant molecules can act as voids in the interlayer of semiconductors, degrade the dense packing of side chains of semiconductors [40] or aggregate themselves [41] which could interrupt in-plane crystallization and cause them to act as trap sites. Therefore, surface doping is highly suitable for organic electronics.

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**Table 2.** Electrical characteristics of OSGTs based on small molecular OSCs compared to those of typical OFETs. The p-channel and n-channel characteristics of OFETs were measured at $V_{DS} = -100$ and 100 V, respectively. The p-channel and n-channel characteristics of OSGTs were measured at $V_{DS} = -20$ and 20 V, respectively.

<table>
<thead>
<tr>
<th>Device structure</th>
<th>OSC</th>
<th>$I_{on}/I_{off}$</th>
<th>$V_{th}$ [V]</th>
<th>$\mu_{measured}$ [cm$^2$ V$^{-1}$ s$^{-1}$]</th>
<th>$\mu_{ideal}$ [cm$^2$ V$^{-1}$ s$^{-1}$]</th>
<th>$r$ [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>OFET</td>
<td>pentacene</td>
<td>&gt;10$^6$</td>
<td>-34.1</td>
<td>1.44 ($\pm 0.12$) $\times 10^{-2}$</td>
<td>7.90 $\times 10^{-2}$</td>
<td>54.86</td>
</tr>
<tr>
<td></td>
<td>BPE-PTCDI</td>
<td>&gt;10$^6$</td>
<td>10.8</td>
<td>1.57 ($\pm 0.17$) $\times 10^{-1}$</td>
<td>1.45 $\times 10^{-1}$</td>
<td>92.36</td>
</tr>
<tr>
<td>OSGT</td>
<td>pentacene</td>
<td>&gt;10$^6$</td>
<td>-62.6</td>
<td>3.21 ($\pm 0.47$) $\times 10^{-2}$</td>
<td>9.22 $\times 10^{-3}$</td>
<td>28.72</td>
</tr>
<tr>
<td></td>
<td>BPE-PTCDI</td>
<td>&gt;10$^6$</td>
<td>14.4</td>
<td>7.74 ($\pm 0.75$) $\times 10^{-2}$</td>
<td>2.04 $\times 10^{-2}$</td>
<td>26.36</td>
</tr>
</tbody>
</table>

$a$ On/off current ratio; $b$ Threshold voltage; $c$ Charge carrier mobility; $d$ The standard deviation; $e$ Reliability factor.
Finally, we constructed a 7 × 7 BPE-PTCDI flexible OSGT array using a Parylene-C substrate (Figure 7a–d). First, 10 μm-thick Parylene-C was coated on an OTS-treated Si wafer (SiO$_2$ 300 nm), and the delaminated Parylene-C layer was used as the actual supporting substrate. Then, Cr/Au (4 nm/40 nm) were thermally evaporated consecutively under high vacuum (≈1.0 × 10$^{-6}$ Torr) for the gate electrodes. A 500 nm thick Parylene-C with a capacitance of 7 nF cm$^{-2}$ was used as a dielectric layer for flexible OSGTs. Then, a 40 nm thick BPE-PTCDI active layer was deposited, as described in the Experimental Section. Au for the source electrode and Al for the drain electrode were thermally evaporated through a shadow mask.

Figure 5. Electrical characteristics of n-type doped N2200 OSGTs compared to typical OFETs. a) Transfer curves of N2200 OSGTs according to n-type dopant concentration (inset: OSGT structure). As the concentration of rPyB increased, the value of $V_{th}$ shifted negatively. b) Output curves of N2200 OSGTs doped with 0.7 mg mL$^{-1}$ rPyB (inset: $I_{DS}$–$V_{DS}$ curves that ranged from 0 to 20 V of $V_{DS}$). c) Transfer curves of N2200 OFETs according to n-type dopant concentration (inset: OFET structure). As the concentration of rPyB increased, the on/off ratio decreased. d) Output curves of N2200 OFETs without rPyB doping. e) $P_{sat}$ changes according to the n-type dopant concentration in OSGTs and typical OFETs. f) $\Delta N_t$ changes according to the n-type dopant concentration in OSGTs and typical OFETs (inset: $V_{th}$ changes).

2.4. Flexible and Low-Power Devices Based on OSGTs
mask. The channel width and length were 440 and 110 μm, respectively, as shown in Figure 7e. Finally, the OSGT devices were peeled off from the Si wafer. The size of the device on the Parylene-C substrate was 2.5 × 2.5 cm². As shown in Figure 7e, the device exhibited highly transparent characteristics such that the background could be seen. The flexible devices exhibited fast saturation behaviors at a low $V_{DS} (\approx 20 \text{ V})$ and stable charge carrier mobilities that were maintained up to 94.6% of the intrinsic mobility after the bending test at a bending radius of 5.8 mm (Figure 7f). The mechanically deformable nature of Parylene-C and BPE-PTCDI combined with OSGT device engineering enabled these flexible and low-power organic electronic devices. Therefore, our results demonstrate the high potential to commercialize low-power flexible devices. In our results, the OSGT s have a low threshold voltage ($V_{th}$) and a low saturation drain voltage ($V_{DS,sat}$) for operation. Furthermore, we fabricated low-power flexible electronic devices based on plastic substrates using low-temperature processes.

### Table 3. Electrical characteristics of N2200-based OSGTs compared to those of typical OFETs. The characteristics of OFETs were measured at $V_{DS} = 100 \text{ V}$. The characteristics of OSGTs were measured at $V_{DS} = 10 \text{ V}$.

<table>
<thead>
<tr>
<th>Device structure</th>
<th>$C_{PyB}$</th>
<th>$I_{on}/I_{off}$</th>
<th>$V_{th}$ [V]</th>
<th>$\mu_e$ [cm² V⁻¹ s⁻¹]</th>
</tr>
</thead>
<tbody>
<tr>
<td>OFET</td>
<td>No doping</td>
<td>$10^4 - 10^5$</td>
<td>12.3</td>
<td>7.29 (±0.33) x 10⁻²</td>
</tr>
<tr>
<td></td>
<td>0.5 mg mL⁻¹</td>
<td>$10^4 - 10^5$</td>
<td>2.0</td>
<td>9.02 (±0.27) x 10⁻²</td>
</tr>
<tr>
<td></td>
<td>0.7 mg mL⁻¹</td>
<td>$10^4 - 10^5$</td>
<td>−23.2</td>
<td>9.53 (±0.64) x 10⁻²</td>
</tr>
<tr>
<td></td>
<td>1.0 mg mL⁻¹</td>
<td>$10^4 - 10^5$</td>
<td>−24.3</td>
<td>4.85 (±0.87) x 10⁻¹</td>
</tr>
<tr>
<td>OSGT</td>
<td>No doping</td>
<td>$10^4 - 10^5$</td>
<td>51.2</td>
<td>1.44 (±0.10) x 10⁻²</td>
</tr>
<tr>
<td></td>
<td>0.5 mg mL⁻¹</td>
<td>$10^4 - 10^5$</td>
<td>13.4</td>
<td>2.09 (±0.15) x 10⁻²</td>
</tr>
<tr>
<td></td>
<td>0.7 mg mL⁻¹</td>
<td>$10^4 - 10^5$</td>
<td>0.1</td>
<td>3.29 (±0.47) x 10⁻²</td>
</tr>
<tr>
<td></td>
<td>1.0 mg mL⁻¹</td>
<td>$10^4 - 10^5$</td>
<td>−55.7</td>
<td>1.63 (±0.20) x 10⁻²</td>
</tr>
</tbody>
</table>

*a) Concentration of rPyB; b) The standard deviation.

Figure 6. Air-stability analyses of rPyB-doped OSGTs (insets (a) and (b)): a) Transfer curves of N2200 OSGTs doped with 0.7 mg mL⁻¹ rPyB as fabricated and after being stored in air for up to 40 d. b) Changes in the electron mobility of N2200 OSGTs under ambient conditions for 40 d. Energy level analyses of rPyB-doped N2200 by UPS and UV–vis spectroscopy (from insets (c)–(e)): c) N2200. d) 0.7 mg mL⁻¹ rPyB-doped N2200. e) Comparison of energy levels of pristine N2200, rPyB-doped N2200 films, Al and Au.
3. Conclusion

Low-voltage operative OSGTs were fabricated using OSCs deposited at a low temperature (<150 °C) as the active layer. The OSGTs were fabricated in a TCBG configuration using asymmetric metal electrodes for the source and drain contacts: Al and Au (or Se). OSGTs showed the manipulating ability of the Schottky barrier using the gate electric field. The mechanism of lowering the Schottky barrier was characterized by activation energy measurements across the temperature range from 298 to 368 K. When increasing the gate voltage from 0 to 10 V in N2200 OSGTs, the Schottky barrier was reduced from 0.24 to 0.16 eV. The OSGTs showed fast \( I_{DS} - V_{DS} \) saturation characteristics compared to typical OFETs, rendering them suitable for low-power applications. The OSGTs based on four representative OSCs, including small molecules and polymeric molecules, exhibited a similar lowered saturation voltage effect. Typical OFETs had a saturation point \( |V_{DS,sat}| > 80 \) V. However, the OSGTs showed low voltage saturation characteristics with \( |V_{DS,sat}| < 20 \) V and well-resolved linear and saturation regimes. Furthermore, to lower the value of \( V_{th} \) for OSGTs, a solution-processed n-type organic dopant, rPyB, was used. This dopant filled the trap sites in the active layer and lowered \( V_{th} \) from 51.2 to 0.1 eV. Using n-type doping, the electron mobilities of n-type doped OSGTs were maintained up to 93.2% for 40 d in an ambient environment. Finally, we made flexible OSGTs based on a Parylene-C substrate that maintained 94.6% electronic mobility, even after bending at a radius of 5.8 mm. This work provides a simple and effective fabrication approach for low-voltage driven electronics. This feature gives an advantage of saving electrical energy, which makes flexible OSGTs suitable for next-generation organic electronics such as wearable devices.

4. Experimental Section

Materials and Instrumentation: All materials were purchased from Aldrich, Alfa Aesar, Lumtec or Polyera and used without further treatment. All solvents were ACS rated unless otherwise noted. ESCALAB 250Xi from Thermo Fisher Scientific was used to measure UPS data under a vacuum of \( 1 \times 10^{-10} \) Torr. The absorption spectra according to wavelength were measured using a Cary 5000 UV–vis–NIR spectrophotometer for thin films. AFM images were obtained using an Agilent 5500 scanning probe microscope operating with a Nanoscope V.
controller. AFM images were recorded in a high-resolution tapping mode under ambient conditions.

Fabrication of OFETs and OSCs: pentacene was thermally deposited under a high vacuum (<5 × 10⁻⁶ Torr) at a speed of 0.01–0.02 nm s⁻¹. The substrate temperature was 65 °C. BPE-PTCDI was thermally deposited at the same speed. The substrate temperature was 125 °C. PDBT-co-PT was dissolved in chlorobenzene at 1.0 mg mL⁻¹. The solution was spin-coated on an OTS treated SiO₂/n-Si substrate at 5000 rpm in an N₂ environment. The PDBT-co-PT film was heat-treated on a hot plate at 130 °C. BPE-PTCDI was thermally deposited at the same concentration and coating conditions. For OFET fabrication, Au and Al electrodes were thermally deposited using shadow masks under a high vacuum (<5 × 10⁻⁶ Torr). For OSC fabrication, Au (or Se) and Al electrodes were thermally deposited using shadow masks.

Preparation of rPyB: PyB, Pyronin B, (0.5, 2.5, 3.5, or 5 mg) was dissolved in 5 mL deionized water, then 5 mL hexane was added to make a two-phase solution. NaBH₄ was added to the two-phase solution so that the molar ratio of NaBH₄ to PyB was 200:1. The resulting two-phase solution was left for 1 d. The deep purple of PyB in deionized water became clear after the reaction. The hexane phase including rPyB was carefully separated using a pipette.

n-Type Doping: For n-type doping, the rPyB solution was spin-coated on the devices at 2000 rpm in an N₂ environment. The devices were not heat-treated because rPyB can provide n-type doping effect at room temperature.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

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