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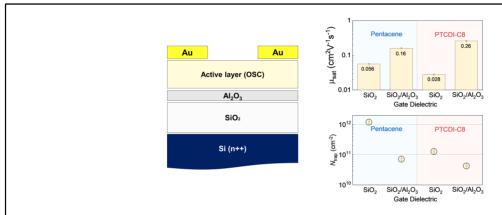
## **RESEARCH**

# Low-Voltage n- and p-Channel Organic Transistors using Al<sub>2</sub>O<sub>3</sub> Dielectric

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### **ABSTRACT**

The integration of high-k dielectric materials presents a promising strategy to reduce the operating voltage of organic thin-film transistors (OTFTs) while enhancing their overall performance. In this study, we explore the impact of a 20 nm Al<sub>2</sub>O<sub>3</sub> interlayer deposited atop conventional SiO<sub>2</sub> dielectrics on the electrical characteristics of both p-channel pentacene and n-channel N,N'-dioctyl-3,4,9,10-perylenedicarboximide (PTCDI-C8) OTFTs. Devices incorporating the Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub> bilayer dielectric demonstrate marked improvements in electrical properties for both n-type and p-type devices, including increased drain current, transconductance, and carrier mobility, along with higher on/off current ratios at reduced operating voltages compared to those using SiO<sub>2</sub> alone. Furthermore, this bilayer architecture lowers threshold and turn-on voltages, decreases subthreshold swing, and mitigates interfacial trap states and contact resistance, thereby promoting more efficient charge injection and enhanced device operation across both charge carrier types. These findings highlight the effectiveness of Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub> dielectric engineering as a viable approach for advancing low-power, high-performance organic electronics.

Key Words: OTFTs, High-k dielectric, Low voltage, Interface trap state, Contact resistance

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#### 1. INTRODUCTION

Organic thin-film transistors (OTFTs) have attracted significant attention as promising candidates for next-generation flexible, low-cost, and large-area electronic applications [1-8]. However, achieving low operating voltages and high performance simultaneously remains a critical challenge, largely limited by the dielectric layer's properties and the organic semiconductor/dielectric interface [9-12]. Furthermore, the interfacial properties between the organic semiconductor and dielectric critically influence charge transport and overall device performance. Engineering and optimizing these interfaces are essential to realize high-performance OTFTs with stable and efficient operation [9-12]. Extensive efforts have been devoted to enhancing OTFT performance, particularly by achieving low-voltage operation through diverse processing strategies [13-24]. Conventional low-k dielectric materials such as silicon dioxide (SiO<sub>2</sub>), with a dielectric constant of ~3.9 [18,24], inherently limit gate capacitance at a given thickness, thereby requiring high operating voltages that are unsuitable for low-power applications. On the other hand, integrating high-k dielectrics such as aluminum oxide (Al<sub>2</sub>O<sub>3</sub>), with a dielectric constant between 6 and 9 [18, 24], provides a promising approach to overcome these limitations by increasing gate capacitance, thereby reducing operating voltages and improving charge modulation [18,24]. While high-k Al<sub>2</sub>O<sub>3</sub> is widely used in inorganic electronics [21,22], its application in OTFTs remains limited, particularly in configurations that avoid complex surface treatments. This is largely due to the sensitivity of organic semiconductors to interfacial disorder, which can introduce trap states and degrade device performance. This gap highlights the need to investigate simplified high-k dielectric strategies that ensure favorable interface characteristics while enhancing OTFT performance. In this regard, engineering bilayer dielectric stacks by integrating high-k Al<sub>2</sub>O<sub>3</sub> atop SiO<sub>2</sub> offers a promising route to improve dielectric function and interfacial compatibility without relying on surface modifications or self-assembled monolayers (SAM). This approach may enable high-performance, low-power p- and n-type OTFTs for future flexible and wearable electronics.

Herein, we systematically investigate the effect of integrating a high-k Al<sub>2</sub>O<sub>3</sub> layer into the gate dielectric stack on hole and electron transport in OTFTs. We directly compare devices using conventional low-k SiO<sub>2</sub> and high-k Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub> bilayers, with p-channel pentacene and n-channel N, N'-dioctyl-3,4,9,10-perylenedicarboximide (PTCDI-C8) as the active layers. ll transistors were fabricated and characterized without any surface treatments or self-assembled monolayers (SAM) on the dielectric surface to investigate the intrinsic nature of Al<sub>2</sub>O<sub>3</sub>. Our detailed evaluation of a 20-nm Al<sub>2</sub>O<sub>3</sub> layer shows significant improvement in electrical performance for both charge carrier types. In particular, devices with the Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub> bilayer operate within  $\pm 10$  V and exhibit markedly enhanced key device parameters compared to those with SiO<sub>2</sub>. This dielectric engineering approach offers a clear pathway to low-power, high-performance OTFTs by increasing the dielectric constant and improving the semiconductor/dielectric interface.

#### 2. MATERIALS AND METHODS

All chemicals used in this study were purchased from



Sigma-Aldrich and used without further purification. The schematic cross-section of the bottom-gate, top-contact OTFT structure fabricated in this work is shown in Fig. 1(a). The energy levels, including the Fermi level of Au, and the HOMO and LUMO levels of pentacene and PTCDI-C8, along with the molecular structures of pentacene and PTCDI-C8, are shown Fig. 1(b). OTFTs were fabricated on heavily n-doped Si wafers serving as both the substrate and gate electrode, coated with a 200 nm thermally grown SiO<sub>2</sub> gate dielectric. Prior to organic layer deposition, substrates were ultrasonically cleaned in acetone, isopropanol, and deionized water, followed by UV/ozone treatment (20 min) and nitrogen drying (5 min). Two gate dielectric configurations were employed: a single-layer low-k SiO<sub>2</sub> (200 nm) and a bilayer high-k Al<sub>2</sub>O<sub>3</sub> (20 nm)/low-k SiO<sub>2</sub> (200 nm). For the bilayer structure, a 20 nm Al<sub>2</sub>O<sub>3</sub> layer was deposited by atomic layer deposition (ALD) atop the SiO<sub>2</sub> dielectric, which served as the initial dielectric layer. The ALD process was conducted at 150°C using trimethylaluminum (TMA) as the metal precursor and water vapor as the oxidant.

Depositions proceeded through alternating, self-limiting exposures to TMA and  $H_2O$ , resulting in a growth rate of approximately 1.1 Å per cycle. Pentacene and PTCDI-C8 were thermally evaporated as p- and n-type organic semiconductors, respectively, at 0.05 nm/s under  $5\times10^{-6}$  mbar vacuum, to a thickness of 50 nm. Gold (Au) source and drain (S–D) electrodes (80 nm) were then deposited through a shadow mask, defining a channel length (L) of 50  $\mu$ m and width (W) of 2,000  $\mu$ m. All electrical characterizations of the OTFTs were conducted in the dark under vacuum ( $\sim10^{-1}$  mbar) using a Keithley HP 5156 semiconductor characterization system.

#### 3. RESULTS AND DISCUSSION

Table 1 summarizes the dielectric type, dielectric constant, capacitance per unit area, bandgap energy, and thickness of each gate dielectric [18,24]. The equivalent total capacitance of the Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub> bilayer dielectric was calculated using the series capacitance

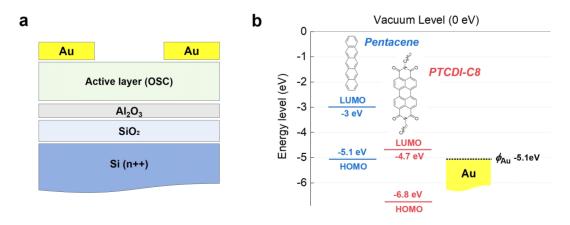


Fig. 1. (a) A schematic cross-section of the bottom-gate, top-contact OTFT structure. (b) Energy level diagram showing the Fermi level of Au and the HOMO/LUMO levels of pentacene and PTCDI-C8, along with the molecular structures of both materials.



**Table 1.** Type, dielectric constant, capacitance per unit area, band gap energy, and thickness of the gate dielectrics used, adapted from Refs. [18,24]

Dielectric materials	k-type	Dielectric constant	Capacitance (nFcm <sup>-2</sup> )	Band gap (eV)	Thickness (nm)
Silicon oxide (SiO <sub>2</sub> )	low-k	3.9	8.3	9	200
Aluminum oxide (Al <sub>2</sub> O <sub>3</sub> )	high-k	9	165	8.8	20
Al <sub>2</sub> O <sub>3</sub> /SiO <sub>2</sub>	high-k /low-k	_	7.9	_	220

model according to:

$$C_{eq} = \frac{1}{C_{Al_2O_3}} + \frac{1}{C_{SiO_2}} \tag{1}$$

where the individual areal capacitances of  $Al_2O_3$  and  $SiO_2$  are given in Table 1. Based on this relation, the equivalent capacitance  $C_{eq}$  of the bilayer dielectric was calculated to be approximately 7.9 nFcm<sup>-2</sup>.

Fig. 2(a)–(b) shows the transfer characteristics ( $I_d$ – $V_g$ ) and transconductance ( $g_m$ ) as a function of  $V_g$  in the linear regime for p-channel ( $V_d$ =–10 V) and n-channel ( $V_d$ =+10V) OTFTs with SiO<sub>2</sub> gate dielectric, respectively. Meanwhile, Fig. 2(c)–(d) displays the corresponding characteristics for p-channel ( $V_d$ =–1 V) and n-channel ( $V_d$ =+1 V) OTFTs with Al<sub>2</sub>O<sub>3</sub> (20 nm)/SiO<sub>2</sub> (200 nm) bilayer gate dielectric, respectively. As shown in Fig. 2(a)–(d), a clear difference in device performance is observed between p- and n-channel OTFTs with SiO<sub>2</sub> (200 nm) and those with Al<sub>2</sub>O<sub>3</sub> (20 nm)/SiO<sub>2</sub> (200 nm) bilayer dielectric.

Devices with  $SiO_2$  required higher operating voltages and exhibited lower drain currents. In contrast, incorporating a 20 nm  $Al_2O_3$  interlayer significantly enhanced the drain current, reaching-1.55×10<sup>-5</sup> A for p-channel and  $2.37\times10^{-5}$  A for n-channel devices at relatively low voltages.

Transconductance  $(g_m)$ , which quantifies the modulation of  $I_d$  as a function of  $V_g$  at a constant  $V_d$ , is

calculated using [25,26]:

$$g_{m} = \left[\frac{\delta I_{D}}{\delta V_{G}}\right]_{V_{D=de}} = \frac{W}{L} \mu linC_{i}V_{d}$$
(2)

where  $\mu_{lin}$  is the linear mobility,  $C_i$  is the capacitance of the insulator per unit area, and W and L represent the channel width and length, respectively.

To ensure a fair device performance comparison, the extrapolation in the linear region (ELR) method [27,28] was employed to extract the maximum drain current ( $I_{d,max}$ ), the gate voltage at peak transconductance ( $V_{g,max}$ ), and the maximum transconductance ( $g_{m,max}$ ) from the transfer and transconductance characteristics in the linear regime of each device, as illustrated in Fig. 2(c). The extracted values of  $V_{g,max}$  and  $g_{m,max}$  are plotted as a function of  $I_{d,max}$  in Fig. 2(e)–(f). Interestingly, both p-and n-channel OTFTs with a  $Al_2O_3/SiO_2$  dielectric exhibited significantly higher  $g_{m,max}$  values,  $1.2 \times 10^{-6}$  S (p-channel) and  $8.6 \times 10^{-7}$  S (n-channel), under low  $V_{g,max}$ , compared to  $5.2 \times 10^{-8}$  S and  $5.4 \times 10^{-7}$  S, respectively, for devices with  $SiO_2$ , confirming the enhanced performance.

These behaviors are mainly attributed to the enhanced charge storage capability at the conductive channel/dielectric interface, where the  $Al_2O_3/SiO_2$  bilayer enables efficient charge carrier accumulation at low operating voltages, leading to increased  $I_d$  and  $g_m$ .

Furthermore, the measured g<sub>m</sub> increases quasi-linearly



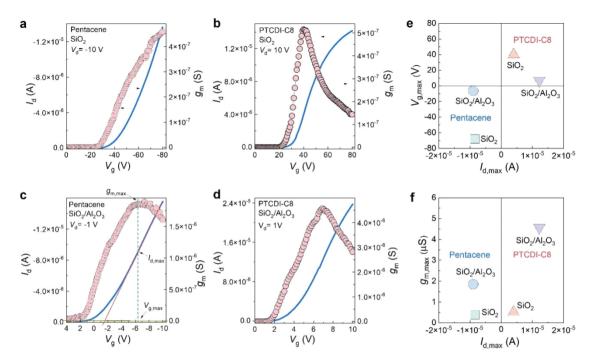


Fig. 2. (a,b) Transfer characteristics  $(I_d-V_g)$  and transconductance  $(g_m)$  as a function of gate voltage  $(V_g)$  in the linear regime for p-channel  $(V_d=-10\ V)$  and n-channel  $(V_d=+10\ V)$  OTFTs with a SiO<sub>2</sub> gate dielectric. (c,d) Corresponding characteristics for p-channel  $(V_d=-1\ V)$  and n-channel  $(V_d=+1\ V)$  OTFTs with an Al<sub>2</sub>O<sub>3</sub> (20 nm)/SiO<sub>2</sub> (200 nm) bilayer gate dielectric. (e,f) extracted  $V_{g,max}$  and  $g_{m,max}$  values plotted as a function of  $I_{d,max}$ .

with  $V_g$  and decreases at high  $V_g$ , as can be seen in Fig. 2(b)–(d). Presumably, the decrease in  $g_m$  at high  $V_g$  results from the presence of contact resistance ( $R_c$ ) at the S–D electrodes/organic semiconductor interface [25, 29].

Fig. 3(a)–(d) presents the transfer characteristics ( $I_d$ – $V_g$ , left axis) and the square root of the drain current versus gate voltage ( $-V_g$ , right axis) in the saturation regime for both p-channel and n-channel OTFTs. The comparison includes devices with a  $SiO_2$  dielectric operated at  $\pm 80$  V and those with an  $Al_2O_3/SiO_2$  bilayer dielectric operated at  $\pm 10$  V. As shown in Fig. 3(c)–(d), both p-channel and n-channel OTFTs incorporating the  $Al_2O_3/SiO_2$  bilayer operate reliably at a low voltage of  $\pm 10$  V, attributed to the high dielectric constant of the

inserted  $Al_2O_3$  layer. In contrast, devices with  $SiO_2$  alone require a higher operating voltage ( $\pm 80~V$ ) to achieve similar electrical performance. A maximum  $I_d$  level of  $-3\times10^{-5}~A$  at -10~V was achieved for the p-channel OTFT, while the n-channel device exhibited  $3.75\times10^{-5}~A$  at +10~V, both employing the  $Al_2O_3/SiO_2$  bilayer dielectric. In contrast, at the same operating gate voltages (-10~V for p-channel and +10~V for n-channel), devices with  $SiO_2$  exhibited significantly lower drain currents of  $-2.6\times10^{-5}~A$  and  $1.84\times10^{-5}~A$ , respectively.

It can be concluded that the use of an Al<sub>2</sub>O<sub>3</sub> interlayer on SiO<sub>2</sub> effectively enables high saturation current operation at low voltage for both p-channel and n-channel OTFTs, outperforming devices with SiO<sub>2</sub>.



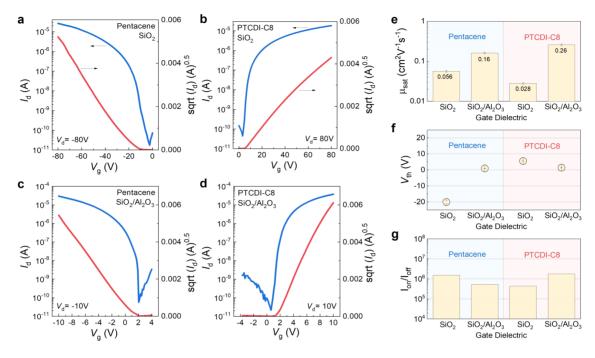


Fig. 3. (a–d) Transfer characteristics ( $I_d$ – $V_g$ , left axis) and  $\sqrt{I_d}$ – $V_g$  plots (right axis) in the saturation regime for p-channel and n-channel OTFTs. Corresponding extracted values of: (e) Maximum saturation mobility ( $\mu_{sat,max}$ ). (f) Threshold voltage ( $V_{th}$ ). (g) Current on/off ratios ( $I_{on}/I_{off}$ ).

In the saturation regime, the mobility is extracted using the following equation [25,26]:

$$\mu_{sat} = \frac{2L}{WC_i} \left( \frac{\partial \sqrt{I_d}}{\partial V_g} \right)^2 \tag{3}$$

The threshold voltage  $(V_{th})$  for each device was extracted by linearly fitting the square root of the transfer characteristics  $(\sqrt{I_d}-V_g)$  in the saturation regime and extrapolating to zero drain current.

The maximum saturation mobility values ( $\mu_{sat, max}$ ) for all devices are shown in Fig. 3(e), while the obtained  $V_{th}$  values are presented in Fig. 3(f). As shown in Fig. 3(e), the highest mobility values were obtained from devices with the  $Al_2O_3/SiO_2$  bilayer dielectric, reaching 0.26 cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup> for n-channel and 0.16 cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup> for p-channel OTFTs, compared

to 0.028 and 0.056 cm<sup>2</sup>  $V^{-1}$  s<sup>-1</sup>, respectively, for devices with SiO<sub>2</sub>.

Devices employing a  $Al_2O_3/SiO_2$  bilayer dielectric exhibit near zero  $V_{th}$ , measured at approximately 1.0 V for p-channel and 1.46 V for n-channel OTFTs, in contrast to devices with a  $SiO_2$  dielectric, which show significantly higher  $V_{th}$  values of -20 V and 5.6 V, respectively. Such low  $V_{th}$  is crucial for the integration of OTFTs into low-voltage circuits [30]. The observed difference in  $V_{th}$  between  $SiO_2$ -based and  $Al_2O_3/SiO_2$ -based devices underscores the strong influence of dielectric composition on device behavior.

The current on/off ratios ( $I_{on}/I_{off}$ ) for all devices are also extracted from the  $I_d$ – $V_g$  characteristics in the saturation regime and are presented in Fig. 3(g). Among all devices studied, the n-channel transistor



with a 20-nm  $Al_2O_3$  insertion layer exhibits the highest  $I_{on}/I_{off}$  ratio of  $1.736\times10^6$ , significantly improved compared to the device with  $SiO_2$ , which shows a ratio of  $4.263\times10^5$ .

In contrast, the p-channel device shows a decrease in  $I_{on}/I_{off}$  when  $Al_2O_3$  is incorporated atop  $SiO_2$ .

Next, the turn-on voltage  $(V_{on})$  refers to the gate voltage at which a conductive channel begins to form between the S-D electrodes [31]. Fig. 4(a) shows the measured values of  $V_{on}$  for all devices. Remarkably, all the  $V_{on}$  values were notably low, suggesting a better gate voltage modulation. Therefore, the low  $V_{on}$  values observed in our p- and n-channel OTFTs are advantageous and comparatively lower than those previously reported in other OTFTs [28,32].

The enhanced performance of both p- and n-channel OTFTs is attributed to the improved interfacial properties introduced by the high-k Al<sub>2</sub>O<sub>3</sub> layer, along

with the advantage of its high dielectric constant, which enhances electrostatic control and enables lower operating voltages. This high-k layer likely reduces the interfacial trap density, resulting in higher drain current, enhanced saturation mobility, and reduced  $V_{\rm th}$ , and  $V_{\rm on}$ . To further investigate this effect, interface trap densities were systematically extracted for all devices across both lower and higher energy regions.

In OTFT devices, the trap density at the dielectric/OSC interface, along with structural disorder in the semiconductor, is often suggested to influence the  $V_{\rm th}$ ,  $V_{\rm on}$ , and SS [33,34]. Here, two independent approaches were employed to estimate the trap densities associated with each dielectric layer.

The first approach estimates the interfacial trap density  $(N_{tr})$  based on the measured  $V_{th}$  and  $V_{on}$ , providing insight into trap-related interfacial characteristics, as given by the following equation [35]:

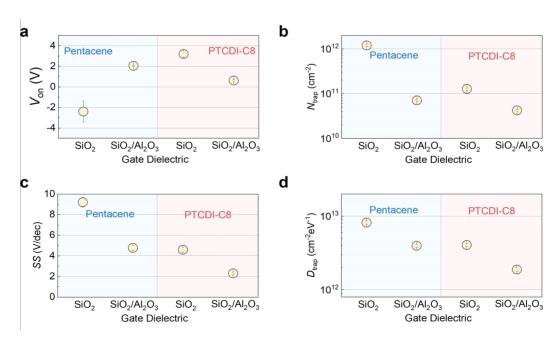


Fig. 4. Extracted values of: (a) Turn-on voltage (V<sub>on</sub>). (b) Trap density (N<sub>tr</sub>). (c) Subthreshold swing (SS). (d) Interface trap density (D<sub>it</sub>) of p-channel and n-channel OTFTs.



$$N_{tr} = \frac{C_i \left| V_{th} - V_{on} \right|}{q} \tag{4}$$

The calculated  $N_{\rm tr}$  values are presented in Fig. 4(b). Notably, the highest extracted trap state densities ( $N_{\rm tr}$ ) were  $1.2\times10^{12}$  cm<sup>-2</sup> for p-channel and  $1.275\times10^{11}$  cm<sup>-2</sup> for n-channel OTFTs employing SiO<sub>2</sub> as the gate dielectric. In contrast, the lowest  $N_{\rm tr}$  values were  $7\times10^{10}$  cm<sup>-2</sup> for p-channel and  $4.23\times10^{10}$  cm<sup>-2</sup> for n-channel devices with the Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub> bilayer. These results indicate that both p- and n-channel OTFTs with Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub> dielectrics exhibit low trap state densities near to the Fermi level [36], corresponding to the low-energy region.

In this second approach, the interface trap density  $(D_{it})$  is extracted from the SS values using the following expression [29, 35]:

$$D_{it} = \left[\frac{SSlog(e)}{kT/q} - 1\right] \frac{C_i}{q} \tag{5}$$

where T is the temperature, k is Boltzmann's constant, q is the elementary charge, and SS is the subthreshold swing. SS serves as a key indicator of device performance and is directly influenced by the trap density at the organic semiconductor (OSC)/gate dielectric interface. It can be extracted from the transfer characteristics using [29]:

$$SS = \frac{dV_g}{dlog(I_d)} \tag{6}$$

The calculated SS and D<sub>it</sub> values are presented in Fig. 4(c), Fig. 4(d), respectively. Although the second method also examines trap states near the Fermi level, it is mainly used to extract the trap state density per unit energy at higher energy levels beyond the range covered by the first method. Fig. 4(d) shows relatively

high interface trap densities at higher energy levels, with values of  $8.15 \times 10^{12}$  cm<sup>-2</sup> eV<sup>-1</sup> (p-channel) and  $4.05 \times 10^{12}$  cm<sup>-2</sup> eV<sup>-1</sup> (n-channel) for SiO<sub>2</sub> devices, which are reduced to  $3.95 \times 10^{12}$  cm<sup>-2</sup> eV<sup>-1</sup> and  $1.88 \times 10^{12}$  cm<sup>-2</sup> eV<sup>-1</sup>, respectively, in devices using the Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub> bilayer. These findings indicate that the differences in electrical performance are primarily influenced by deep trap states located closer to the highest occupied molecular orbital (HOMO) in p-channel devices and the lowest unoccupied molecular orbital (LUMO) in n-channel devices, suggesting that trap states in the high-energy region have a greater impact than those in the low-energy region.

Fig. 5(a), Fig. 5(b) shows the output characteristics (I<sub>d</sub>-V<sub>d</sub>) of p-channel and n-channel OTFTs with a SiO<sub>2</sub> gate dielectric, while Fig. 5(c), Fig. 5(d) displays the corresponding characteristics of devices employing an Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub> bilayer dielectric. As shown in Fig. 5(c), Fig. 5(d), the incorporation of a 20 nm high-k Al<sub>2</sub>O<sub>3</sub> layer enables both p- and n-channel OTFTs to operate at low voltages (±10 V), compared to the higher voltage requirements of devices using SiO2. All devices exhibit a clear transition from the linear to the saturation regime, where I<sub>d</sub> increases linearly at low V<sub>d</sub> and saturates at higher V<sub>d</sub> due to channel pinch-off. The introduction of the Al<sub>2</sub>O<sub>3</sub> layer enhances the I<sub>d</sub> level at low applied drain and gate voltages, as evidenced by the output characteristics. This result further confirms that the Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub> bilayer dielectric improves the semiconductor/dielectric interface, facilitating more efficient charge transport and leading to higher I<sub>d</sub> levels.

R<sub>c</sub> can significantly hinder charge injection, thereby limiting the overall performance of OTFTs [28,37,38]. Understanding its impact on carrier injection from the S–D electrodes to the conductive channel is essential



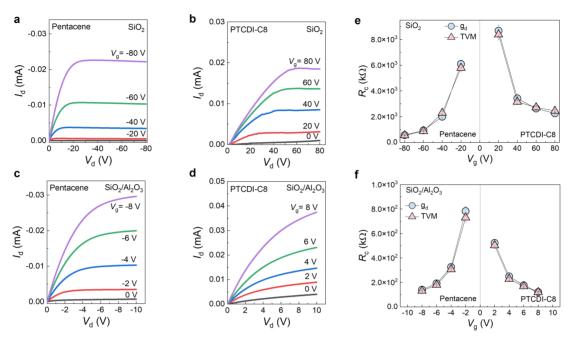


Fig. 5. (a–b) Output characteristics (I<sub>d</sub>–V<sub>d</sub>) of p-channel and n-channel OTFTs with a SiO<sub>2</sub> gate dielectric. (c–d) Corresponding characteristics of devices with an Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub> bilayer gate dielectric. (e–f) Contact resistance (R<sub>c</sub>) values extracted using both methods as a function of V<sub>g</sub>.

for optimizing device efficiency. Several methods have been developed to extract  $R_{\rm c}$ , including the Y-function method, the transfer line method (TLM), the transition voltage method (TVM), and the output conductance method [28,37,38]. Herein, the influence of the dielectric on contact resistance was assessed using two complementary methods: the transition voltage method (TVM) and the conductance method, both of which were employed to accurately extract  $R_{\rm c}$  of the investigated OTFTs.

At low drain voltage, the output conductance of the devices can be expressed as follows [39]:

$$g_d = \frac{1}{R_{tot}} = \left[\frac{\delta I_d}{\delta V_d}\right] V_{g=cte} \tag{7}$$

For each device and at each  $V_g$ , the total source-drain resistance ( $R_{tot}$ ) is extracted from the slope of the

linear region of the  $I_d$ - $V_d$  output curve, using  $R_{tot} = \left[\frac{\delta V_d}{\delta I_d}\right] V_{g-cte}.$ 

 $R_c$  can be extracted as follows:

$$R_{C} = \frac{1}{g_{d}} - \frac{L}{WC_{i}\mu_{FE,lin}(V_{g} - V_{th})}$$
(8)

In addition, an alternative method to extract  $R_c$  is the transition voltage method (TVM), where it is expressed as follows [28]:

$$R_{C} = 2 \frac{\sqrt{V_{g} - V_{th}} \left[ \sqrt{2 V_{tr}} - \sqrt{V_{g} - V_{th}} \right]}{I_{sat}}$$
(9)

Where,  $V_{tr}$  is the transition voltage from linear to saturation regime in the output characteristics, and  $I_{sat}$  is the saturation drain current. At a given  $V_{\rm g}$ ,  $V_{\rm d} = V_{\rm tr}$  and  $I_{lin} = I_{sat}$ . The  $V_{\rm tr}$  and  $I_{\rm sat}$  values are derived from the



intersection of the two slopes in the linear and saturation regimes of the output characteristics. The extracted R<sub>c</sub> values of all p- and n-channel OTFTs with SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub> bilayer dielectrics are plotted as a function of V<sub>G</sub> in Fig. 5(e)–(f), respectively. Notably, the R<sub>c</sub> values obtained from both methods show a good agreement, underscoring the reliability of the analysis. as demonstrated in Fig. 5(e)-(f). The extracted R<sub>c</sub> values at high V<sub>G</sub>=±80 V for OTFTs employing SiO<sub>2</sub> as the gate dielectric are  $5.68 \times 10^2$  k $\Omega$  for the p-channel pentacene device and  $2.25\times10^3$  k $\Omega$  for the n-channel PTCDI-C8 device. Upon introducing a 20 nm high-k Al<sub>2</sub>O<sub>3</sub> layer atop the SiO<sub>2</sub> dielectric, R<sub>c</sub> values decrease substantially for both device types. For the p-channel device,  $R_c$  is reduced to  $1.4 \times 10^2$  k $\Omega$ , while the n-channel device shows a further decrease to  $1.2\times10^2$  k $\Omega$ , both measured at V<sub>G</sub>=±8 V. Under these conditions, both devices exhibit comparable R<sub>c</sub> values. The reduction in R<sub>c</sub> observed in devices with Al<sub>2</sub>O<sub>3</sub>/ SiO<sub>2</sub> compared to those with SiO<sub>2</sub> alone is likely attributed to a lower trap density at the semiconductor/ dielectric interface, which promotes more efficient charge carrier transport owing to the beneficial influence of the additional high-k Al<sub>2</sub>O<sub>3</sub> layer.

The comparative analysis of electrical characteristics of both p-channel and n-channel OTFTs with  $SiO_2$  and  $Al_2O_3/SiO_2$  gate dielectrics reveals a consistent and compelling trend of performance enhancement across all metrics, underscoring the effectiveness of the bilayer dielectric strategy. Devices employing the  $Al_2O_3/SiO_2$  bilayer exhibit superior performance, marked by higher  $\mu_{sat}$ , steeper SS, lower trap densities, and reduced  $R_c$  compared to their  $SiO_2$  counterparts. These improvements are mainly attributed to the dual benefits of the

high-k Al<sub>2</sub>O<sub>3</sub> layer: (i) enhanced electrostatic control due to the increased dielectric constant, which enables efficient charge carrier accumulation at low operating voltages, and (ii) a reduction in interface trap states at the semiconductor–dielectric interface, facilitating more efficient charge transport. Importantly, these enhancements were achieved without any surface treatments or SAMs, underscoring the intrinsic ability of high-k Al<sub>2</sub>O<sub>3</sub> to form a favorable and trap-minimized interface with organic active layers. These results further underscore the effectiveness of this dielectric engineering strategy in advancing low-power, high-performance organic electronics.

#### 4. CONCLUSION

In summary, p-channel pentacene and n-channel PTCDI-C8 OTFTs were fabricated using either a SiO<sub>2</sub> dielectric or Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub> bilayer. The bilayer dielectric effectively lowers the operating voltage to  $\pm 10$  V while improving charge transport, thereby significantly enhancing overall device performance. Devices with Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub> show improved mobility (0.16 cm<sup>2</sup>V<sup>-1</sup>s<sup>-1</sup> for holes and 0.26 cm<sup>2</sup>V<sup>-1</sup>s<sup>-1</sup> for electrons), higher transconductance, near-zero V<sub>th</sub> and V<sub>on</sub>, and higher on/off current ratios. The Al<sub>2</sub>O<sub>3</sub> layer also significantly lowers  $R_c$  (1.4×10<sup>2</sup> k $\Omega$  for p-channel and 1.2×10<sup>2</sup> k $\Omega$ for n-channel) and trap densities. These improvements are mainly attributed to the dual benefits of the high-k Al<sub>2</sub>O<sub>3</sub> layer, which enhances electrostatic control through its higher dielectric constant, enabling lower operating voltages, and simultaneously reduces interface trap states at the semiconductor/dielectric interface, facilitating more efficient charge transport. Together, these findings highlight the importance of



dielectric interface engineering for improving both pand n-type OTFT performance and enabling energyefficient, low-power organic electronics.

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#### **Author Contributions**

WB performed the experiment and wrote manuscript. SW, CLT and HS were involved in analysis, and discussion. DK wrote the manuscript and supervised this work. All authors read and approved the final manuscript.

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#### **Declarations of Competing Interests**

The authors declare that they have no competing interests.

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