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Impact of the Gate Dielectric on Contact Resistance and Bias-Stress Stability of High Mobility Organic Transistors

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Abstract

The impact of the gate dielectric on contact resistance in organic thin-film transistors (OTFTs) is investigated using electrical characterisation, bias-stress stability measurements, and bandgap density of states (DOS) analysis. Two similar dielectric materials namely, Cytop and poly[4,5-difluoro-2,2-bis(trifluoromethyl)-1,3-dioxole-*co*-tetrafluoroethylene] (Teflon™ AF2400), are tested in top-gate bottom-contact OTFTs. The contact resistance of Cytop-based OTFTs is found to be greater than that of the AF2400-based devices, even though the metal/OSC interface remains identical in both systems. The Cytop devices are also found to perform worse in the bias-stress stability tests which, along with the DOS calculations, suggests that charge trapping at the OSC/dielectric interface is more prevalent with Cytop than AF2400. This increased charge trapping at the Cytop OSC/dielectric interface appears to be associated with the higher contact resistance in Cytop OTFTs. Differences in the molecular structure between Cytop and AF2400 and the large difference in the glass transition temperature of the two polymers may well be responsible for the observed difference in the transistor performance. Overall, this study highlights the importance of the gate dielectric material in the quest for better performing OTFTs and integrated circuits.

1. Introduction

Organic thin-film transistors (OTFTs) have attracted great interest over the last 30 years in the quest to achieve flexible, low cost, large-area, printable electronics. Worldwide research has led to remarkable improvements in OTFT operating parameters, with the greatest focus placed on improving the charge-carrier mobility, μ .^{[1] [2] [3] [4] [5]} However, these successful improvements in μ will be in vain unless other important operating parameters are also improved. One particularly

weak point in OTFTs that use undoped, wide-bandgap organic semiconductors (OSC) is their contact resistance (R_C). Not only are the μ values reported for OTFTs suffering from R_C at a high risk of being overestimated or underestimated,^{[1] [6] [7]} but a large R_C can also prevent OTFT miniaturization.^[8] These points are important if OTFTs are ever to be used in integrated circuits (ICs), where IC operating frequencies can be increased by reducing the channel length and improving μ .^{[9] [10]} Indeed, Hagen Klauk recently calculated two key requirements for OTFTs to be used in GHz frequency applications for the “internet of things”: (i) R_C less than 100 Ωcm , and (ii) channel dimensions either equal to or less than 1 μm .^[9] In principle, these requirements be met by improving R_C .

Although R_C is extracted as a single value from the OTFT output characteristics, its magnitude encompasses different aspects of OTFT operation. The largest contributor to R_C is the potential energy difference between the metal contact and the OSC. In OTFTs, this energetic difference typically creates a Schottky barrier, which reduces how efficiently charge carriers are injected into/extracted from the OTFT channel.^{[11] [12]} Therefore, much research has focused on techniques that improve this energetic mismatch, such as contact doping,^{[13] [14] [15]} alternative contact materials, OSC doping,^{[4] [16] [17]} injection layers,^{[18] [19]} and self-assembled monolayer (SAM) treatments.^{[20] [21]} However, there are a number of additional factors that influence the magnitude of R_C , including the OSC microstructure,^{[22] [23]} charge trapping at the metal/OSC interface,^[24] as well as the type of dielectric materials employed^{[25] [26]} and the microstructure they create at the OSC/dielectric interface.

The dielectric layer in OTFTs has attracted significant attention over the years for a variety of reasons. For instance, the all-important charge accumulated channel forms within a few nanometers of the OSC/dielectric interface.^{[27] [28]} Also, the dielectric influences numerous OTFT

parameters, such as operating voltage,^[29] threshold voltage,^{[30] [31]} on-off current,^[32] subthreshold swing/slope,^[33] hysteresis,^[34] mobility,^[35] and operational stability.^{[36] [37]} However, the relationship between the dielectric and R_C is rarely considered. At the time of writing, there are only a few studies that either mention or explore how the dielectric impacts R_C in OTFTs.^{[22] [25] [26] [38] [39]} Still, all of these studies utilize bottom-gate top-contact (BG-TC) OTFT architectures and, therefore, may not completely eliminate the influence of OSC ordering and morphology on R_C . Given the significance of R_C in OTFTs, further demonstrations of the relationship between the dielectric and R_C are beneficial for both awareness and resolving the problem of R_C .

Here, we explore the effect of the dielectric layer on R_C in solution-processed OTFTs. We investigated two different but similar fluoropolymer dielectrics, Cytop and poly[4,5-difluoro-2,2-bis(trifluoromethyl)-1,3-dioxole-*co*-tetrafluoroethylene] (Teflon™ AF2400 or AF2400), in top-gate bottom-contact (TG-BC) small-molecule/polymer-blend OTFTs made from C₈-BTBT and C₁₆IDT-BT (more details below). Though the devices were identical except for the material and thickness of the dielectric layer, when R_C was compared at equivalent electric flux densities it was found to be greater in the Cytop OTFTs than in the AF2400 OTFTs. We further explored the hypothesis that charge trapping at the OSC/dielectric interface is responsible for the difference in R_C using bias-stress tests and density of states (DOS) calculations, finding that Cytop OTFTs performed worse than the AF2400 OTFTs in the bias-stress tests, despite the AF2400 being biased under more strenuous conditions. Additionally, density of states analysis suggests that there is less charge carrier trapping in the AF2400 devices than in the Cytop devices. These combined results indicate that charge trapping at the OSC/dielectric interface contributes towards the R_C value and is responsible for the worse R_C of the Cytop devices. Overall, we highlight the importance of the dielectric layer on improving R_C in OTFTs.

2. Results and Discussion

For this study, we employed a solution-processed small-molecule/polymer blend system as the OSC layer in a TG-BC OTFT architecture (**Figure 1a**) using pentafluorothiophenol (PFBT) as the contact workfunction modifier agent (**Figure 1b**). The OSC inks were formulated by blending small-molecule 2,7-dioctyl[1]benzothieno[3,2-b][1]benzothiophene (C₈-BTBT) (**Figure 1c**) with the polymer indacenodithiophene-benzothiadiazole (C₁₆IDT-BT) (**Figure 1d**) to create a C₈-BTBT:C₁₆IDT-BT blend system. We chose this blend for its spin-coating ease and simplicity and its ability to reach extremely high mobilities when incorporated in TG-BC transistors architectures (**Figure 1a**).^{[16] [40] [17]} The latter applicability to TG-BC device architectures is well-known for such solution processed small-molecule/polymer blends because of the vertical phase separation between the two organic components, leaving a layer of high-mobility small-molecule at the surface/air interface and hence the transistor channel.^{[41] [41]} To this end, the use of the TG-BC OTFT architecture is an essential component to this study, ensuring that the semiconducting channel interface with the different dielectrics remain identical. This allowed us to isolate the influence of the semiconductor/dielectric interface on R_C .

We chose two polymeric dielectrics, Cytop (**Figure 1e**) and poly[4,5-difluoro-2,2-bis(trifluoromethyl)-1,3-dioxole-*co*-tetrafluoroethylene] (Teflon™ AF2400 or AF2400) (**Figure 1f**). Cytop is a favourite choice for the dielectric layer in OTFTs, being well-known for its commercial availability, its applicability to both hole and electron transporting materials, low dielectric constant and the fact that it is processed from a solvent that is orthogonal to many OSCs and, therefore, suitable for top-gate OTFT structures.^[42] Similarly, AF2400 is another commercially available polymeric dielectric material that can be processed from orthogonal solvents and also has a low dielectric constant. Although AF2400 is not as widely used as Cytop,

our earlier work showed that AF2400 is a promising dielectric material for OTFTs, particularly for C₈-BTBT:C₁₆IDT-BT blend OTFTs when processed at optimized conditions.^[40] In addition, Cytop and AF2400 have similar permittivity values ($\epsilon_{CYTOP} \approx 2.05$ and $\epsilon_{AF2400} \approx 1.91$) and similar chemical structures that are characterized by the absence of polar OH groups, hence eliminating adverse effects associated with higher permittivity dielectrics^[43] and differing chemical structures.^[25] For the purpose of this study we focused on best performing transistors based on the two dielectrics. Details on the material and device preparation can be found in the *Experimental* section. For each dielectric the capacitance-per-unit area and permittivity were evaluated via capacitance-voltage measurements performed on metal-insulator-metal capacitor structures (**Figure S1**).

2.1. Contact Resistance Analysis

Representative transfer characteristics for the C₈-BTBT:C₁₆IDT-BT blend transistors, both with Cytop and with AF2400, are shown in **Figure 2a-b**, together with the calculated hole mobility values plotted as a function of V_G (**Figure 2c-d**). The corresponding output characteristics are presented in **Figure S2**. The differences between the two devices are instantly visible: compared to AF2400, the Cytop device required larger operating voltages, had a lower drain current (I_D), a much higher threshold voltage, V_T , ($V_{T_Cytop} = -32.8$ V and $V_{T_AF2400} = -9.8$ V), a somewhat more pronounced gate voltage (V_G) dependent mobility once the channel switches on, and a larger subthreshold swing ($SS_{Cytop} = 4.9$ V/decade and $SS_{AF2400} = 2.4$ V/decade; see **Table 1**). The difference in the operating voltages, I_D levels, and (to a certain extent) V_T , can be attributed to the significant difference in the thicknesses of the two dielectrics, which are ≈ 300 nm and ≈ 1100 nm for AF2400 and Cytop, respectively.

On the other hand, the higher V_T , larger SS, and slightly heightened V_G -dependent mobility of the Cytop OTFTs is often attributed to charge trapping at the dielectric/OSC interface^[44] as well as on the semiconductor's structural disorder.^[45] To investigate the root cause of the observed differences, we employed two different methods to calculate the trap densities for the two dielectrics. The first method relies on the experimentally determined values for V_T and onset voltages (V_{on}) and allows estimation of the interfacial hole trap density (N_{tr}) using:

$$N_{tr} = \frac{C_i |V_T - V_{on}|}{q} \quad (1)$$

where C_i is the areal capacitance of the dielectric layer used and q is the elementary charge. Analysis of the data yields N_{tr} values of 2.3×10^{11} and $2.0 \times 10^{11} \text{ cm}^{-2}$ for the Cytop and AF2400 based TFTs (**Table 1**). Interestingly, despite the clear differences in the transistor characteristics, the values for N_{tr} are roughly the same in both systems. This suggests that both systems have a similar number of trap states within this low energy region.

Although the second method also probes traps states within close proximity to the Fermi level, it is often used to estimate the density of states per unit energy (in $\text{cm}^{-2} \text{ eV}^{-1}$) at higher energy levels than Eq. 1. Using this approach, a maximum value for the density of interface traps (D_{tr}^{SS}) can from the SS values measured at $V_G < V_T$ be calculated using^[46]:

$$D_{tr}^{SS} = \frac{C_i}{q} \left(\frac{qSS}{k_B T \ln(10)} - 1 \right) \quad (2)$$

where, k_B is the Boltzmann constant and T is temperature. The D_{tr}^{SS} values calculated using Eq. 2 for Cytop and AF2400, were 8.7×10^{11} and $1.3 \times 10^{12} \text{ cm}^{-2} \text{ eV}^{-1}$, respectively. The combined results

imply that both systems are characterized by a similar number of trap states close to the Fermi level,^[47] suggesting that the observed difference is due to trap states located at higher energies i.e. closer to the highest occupied molecular orbital (HOMO) energy level.

To evaluate the impact of the dielectric on contact resistance we measured OTFTs with a constant channel width (W) and varying channel length (L) so that we could extract R_C for the Cytop and AF2400-based devices using the transmission line method (TLM):^[48]

$$R_{ON} = \frac{\partial V_{D_lin}}{\partial I_{D_lin}} = r_{channel}L + R_C \quad (3)$$

Here, R_{ON} is the total device resistance measured (i.e., drain/source and channel resistance combined) as a function of L , $r_{channel}$ is the resistance across the channel, and R_C is the combined resistances at the source/drain electrodes. **Figure S3** shows the contact resistance ($R_C W$) values, extracted from **Figure S4**, as a function of V_G for the two dielectric materials, showing R_C for the Cytop devices is up to an order of magnitude greater than the AF2400 devices, particularly at low V_G -bias. However, we note that the two dielectric layers, which have been designed to yield OTFTs with optimized performance, are of different thicknesses. The thinner nature of the AF2400 layer, for example, would results to the semiconductor channel being subjected to higher gate fields, which in turn can influence the hole injection and overall charge transport across the channel. In order to compare the two dielectrics fairly, we calculated R_C as a function of electric flux density (D) for the two systems given as:

$$D = C_i \times V_G \quad (4)$$

For the purpose of this analysis we fabricated Cytop and AF2400-based OTFTs with a range of dielectric thicknesses, and hence areal capacitances, to access a broad range of electric flux

densities over which to compare R_C (see **Figure S5**, **Figure S6**, **Table S1** and the description in the *Supporting Information* for further details). The results are presented in **Figure 2e**, demonstrating the same behaviour observed in **Figure S3**, where the R_C value appears greater in Cytop than AF2400-based devices. We do note, however, that as the electric flux density increases (becomes more negative for larger V_G) the difference in R_C between the two dielectrics reduces.

To understand the reasons for the differences in the data in **Figure 2e**, we need to look at all the factors influencing R_C . First, the injection barrier at the metal/OSC interface is the primary contributor to the R_C magnitude in OTFTs.^{[11] [12]} In our experiments, both the Cytop and AF2400 devices rely on identical Au source/drain electrodes (**Figure 1b**) that have been treated with the self-assembled monolayer (SAM) PFBT (**Figure 1b**). The latter was applied to Au electrodes to improve the energetic mismatch between the highest occupied molecular orbital (HOMO) level of C₈-BTBT:C₁₆IDT-BT blend and the workfunction of the Au source/drain electrodes. Therefore, we assume that the injection barriers at the metal/OSC interface remain identical in both systems, and did not cause the differences seen in **Figure 2e**.

The microstructural ordering in the bulk of the OSC layer and at the metal/OSC interface^{[1] [49] [23]} as well as charge trapping at the electrode^[50] is also known to impact R_C . As discussed earlier, we intentionally used TG-BC OTFT architecture to ensure that the dielectric layer did not influence how the blend OSC formed, therefore excluding any influences from OSC morphology. **Figure S7** shows polarized optical microscope images of the C₈-BTBT:C₁₆IDT-BT blend films used in the Cytop and AF2400 OTFTs. There are no discernable differences in the blend OSC morphology; therefore, we assume that the R_C values were not influenced by the microstructure, of the OSC, or the resultant charge trapping at the S-D/OSC interface.

Given the lack of discernable differences in the OSC morphology and metal/OSC interface in the Cytop and AF2400 OTFTs, we disregard the most commonly discussed factors that contribute to R_C . Yet, these OTFT systems, which only differed in their dielectric layers, still produced significantly different R_C values. There are only a handful of studies that discuss the relationship between the gate dielectric and R_C and its potential causes. One example is the work by Blanchet et al., who also discounted any influences from morphology and the metal/OSC interface, and suggested that differences in dielectric chemical nature leads to differences in R_C .^[25] In particular, they noted that R_C was worse in OTFTs fabricated with an acidic dielectric layer than in the OTFTs with a basic dielectric. In our work, we used Cytop and AF2400 that are chemically very similar (**Figure 1e-f**); therefore, we discount the chemical natures as the origin of R_C differences.

Liu et al. published another study that explains the relationship between the dielectric and R_C with accumulated charge trapping at the OSC/dielectric interface.^[26] Trapping at the OSC/dielectric interface is already known to heavily influence device operation,^{[20] [35] [51]} and the nature of the dielectric material can influence the amount of interfacial defects and trap states.^[52] Liu et al. observed a dielectric-dependent R_C in OTFTs that were made with the same OSC and suggested that the reason for this dependency on the dielectric was that different amounts of charge carriers were trapped at the OSC/dielectric interface as they were injected into the channel.^[26] Trapped charge carriers increase R_C by blocking charge injection, screening V_G , and limiting the mobile charge carriers injected from the source into the channel, where the latter reduces the effective carrier mobility.^{[26] [54] [55]} Additionally, others have shown that interfacial layers can be used to reduce charge trapping at the OSC/dielectric interface which in turn reduces R_C .^{[22] [38]}

Therefore, Liu's explanation and other subsequent studies strongly suggest that interfacial charge trapping is responsible for the different R_C that we measure in our Cytop and AF2400 OTFTs.

2.2 Dielectric Influence on Bias-Stress Stability

To gain insight into the trapping mechanisms in our devices and how this may influence R_C , we evaluated their negative bias-stress stability. Bias-stress stability is characterized by a shift in V_T and change in I_D ; these changes are associated with charge trapping whilst the device is in operation and are known to pose a problem in OTFTs.^{[44] [56] [57] [58] [59]} We performed 12-hour (43,200 sec) stability tests on the optimized Cytop and AF2400 C₈-BTBT:C₁₆IDT-BT OTFTs. The OTFTs that we used for these measurements had channel lengths of 80 μm and widths of 1000 μm . Both dielectric systems were biased for 12 h under identical inert nitrogen conditions, with the Cytop devices being biased at $V_{G_Cytop} = -80$ V and the AF2400 OTFTs being biased at $V_{G_AF2400} = -60$ V. We chose to bias the devices with the maximum operating voltages employed during transistor characterization, to keep in-line with the preferred operating conditions of the best-performing devices, although we note that the two systems will be biased under different electric flux densities.

Figure 3 shows the results from the bias-stress tests; the before and after bias-stress transfer characteristics for the Cytop and AF2400 devices are shown in **Figure 3a** and **3b**, respectively. For the Cytop devices, we see that V_T shifted from $V_{T_0h} = -23.6$ V to $V_{T_12h} = -31.6$ V, making the total shift after 12 h of stress $\Delta V_{T_Cytop} = -8$ V. For the AF2400 devices, V_T shifted from $V_{T_0h} = -7.7$ V to $V_{T_12h} = -10.7$ V, making the total shift after 12 h of stress $\Delta V_{T_AF2400} = -3$ V. The density of trap states, Δh^+ , created whilst stressing can be calculated using:

$$\Delta h^+ = \frac{C_i |V_{T_0h} - V_{T_12h}|}{q} \quad (5)$$

where, $\Delta h^+_{\text{Cyttop}} = 9.6 \times 10^{14}$ and $\Delta h^+_{\text{AF2400}} = 3.5 \times 10^{10}$. We can also see the effects of stressing in the before and after transfer curves. The post-stress devices have a slower turn on than the pre-stress devices; this is shown by a flatter region on the transfer curves and the more gradual upwards sweep in $\sqrt{I_D}$ vs. V_G at low voltages. Interestingly, the data suggests that the C₈-BTBT:C₁₆IDT-BT OTFTs with AF2400 are more bias-stable than those based on Cyttop, despite the AF2400 devices being stressed at a significantly higher electric flux density of 3×10^{-7} as compared to 1.4×10^{-7} for Cyttop (D_{Cyttop}).

Figure 3c provides detailed insight into how V_T evolves over time. The two stressed data sets change sharply initially and then plateau with time; this behavior is typical and often explained by the charge-carrier trapping rates reducing over time as the trap states gradually become occupied.^[60] Such behaviour fits with the following stretched exponential function, which can be used for a closer examination of the bias-stress data:^{[2] [61] [60]}

$$\Delta V_T(t) = \Delta V_0 \left\{ 1 - \exp \left[- \left(\frac{t}{\tau} \right)^\beta \right] \right\} \quad (6)$$

where $V_{T_initial}$ is V_T at 0 seconds, $\Delta V_0 = V_G - V_{T_initial}$ (where V_G is the gate voltage applied during stressing), β is the stretching parameter and has a value between 0 and 1, and τ is the relaxation time. The τ and β parameters were extracted from the solid lines in **Figure 3c**. For the Cyttop device, the stretching parameter is $\beta_{\text{Cyttop}} = 0.37$ and the relaxation time is $\tau_{\text{Cyttop}} = 1.26 \times 10^7$ s. For the AF2400 device, the stretching parameter is $\beta_{\text{AF2400}} = 0.36$ and the relaxation time is $\tau_{\text{AF2400}} = 4.52 \times 10^7$ s. The coefficient of determination, R^2 , is 0.996 for the Cyttop devices and 0.994 for the AF2400 devices. The differences between these extrapolated parameters offer insight into the timescales of any trapping that occurred at the two different OSC/dielectric interfaces.

While the two β values are very similar, there is some difference, with $\beta_{\text{Cytop}} > \beta_{\text{AF2400}}$, suggesting that the trap distribution in the AF2400 devices is slightly more uniform.^[62] There is more of a difference between the relaxation times of the two systems, where the AF2400 OTFTs have a higher value of $\tau_{\text{AF2400}} = 4.52 \times 10^7$ s than the Cytop OTFTs at $\tau_{\text{Cytop}} = 1.26 \times 10^7$ s. This implies that the trapping processes occurring in the AF2400 system happen on a smaller timescale and there is a lower probability of charges being trapped.^[22] These results suggest that, overall, charge trapping is worse in Cytop-based than in AF2400-based OTFTs, further supporting the hypothesis that charge trapping at the OSC/dielectric interface is responsible for the increased R_C in the Cytop OTFTs as compared to AF2400-based devices.

2.3 Density of States (DOS) Analysis

To better understand the differences between Cytop and AF2400-based OTFTs in terms of charge trapping, we performed DOS calculations using the Grünwald^[40] model on both unstressed devices (0 h) and devices that had been subjected to continuous bias stress at constant V_D and V_G for 12 h. We determined the density of the immobile states^{[47] [63]} by comparing the field-induced charge with the mobile holes (manifested as I_D) using the experimentally measured transfer characteristics. The Grünwald model consists of three fundamental equations to extract the DOS inside the bandgap as a function of energy with respect to the Fermi level (E_F), when the device is biased at flat-band voltage. A full derivation of the model can be found on Grünwald's original paper,^[63] while a brief overview can be found in our earlier work.^[40] The latter model has already been successfully applied to the C₈-BTBT:C₁₆IDT-BT blend as well as on similar small-molecule/polymer blend-based OTFTs earlier.^{[40] [64]} Therefore, we use the same key approximations that were used for these earlier studies; a justification for these approximations is

given in Hunter *et al.*^[64] We also note that negligible R_C is a Grünwald model assumption and discuss this point further with the results.

Figure 4 shows the results of the DOS analysis. From the differences between the pre-stress devices (bias stress time = 0 h), we see that both systems show similar concentrations of DOS at lower energies. This correlates with the values calculated earlier for N_{tr} and D_{tr}^{SS} , which are roughly the same in both systems. However, at higher energies, the bandgap DOS of the AF2400 system increases much more rapidly than that of the Cytop system. From this, the observed sharper turn-on and steeper subthreshold swing in the transfer characteristics could be anticipated (**Figure S2**). It is often assumed that, in functioning transistors, the high energy super-exponential component of the immobile states is due to the Fermi level approaching the HOMO level, and the model assumptions breaking down.^[64] ^[65] Given this assumption, the data suggests that the energy distance between the E_F and HOMO level is much smaller in devices with AF2400 than Cytop. Additionally, the sharper increase in the bandgap states suggests that there is a lower density of trap states (which can be calculated by integrating under the area of the curve), correlating well with the explanation behind the lower R_C and enhanced bias-stress stability of the AF2400 transistors. After 12 h of stress tests, the DOS of both dielectric systems decreased, though the decrease of the Cytop system was more pronounced, implying that the preexisting trap states were either filled by holes (deactivated) or no trapping/detrapping occurs on the timescale of the transistor measurement during device operation. Although we note that a possible source of error could arise from the model's assumption that R_C is negligible, Kalb *et al.* demonstrated that R_C in OTFTs leads to slight overestimations (increase) of the DOS at energies close to the VB/HOMO.^[66] Therefore, the decrease in DOS that we observe after bias stressing cannot be due to changes in R_C affecting the analysis, allowing us to use the relative shift to infer information about

differences in charge trapping between the two dielectric systems. We also note that if the filling of the traps affected the overall charge balance in the material, then we would expect to see a shift in the DOS instead of a decrease at a certain voltage; we demonstrated such a DOS shift in our earlier work.^[40] We do not observe a clear shift here.

Although the energetic difference at the metal/OSC interface is the primary contributor to the value of R_C ,^[22] we have shown that charge trapping at the OSC/dielectric interface on its own also creates a difference in R_C , even between two dielectrics that are reasonably similar in chemical structure. These fluoropolymers are less polar than some other dielectrics employed in OTFTs, and do not contain OH, which is known to cause trapping problems from ionic impurities and moisture.^{[26] [67] [68]} Interestingly, even though these materials are at the trap-free end of the OTFT-dielectric spectrum, we still see a difference in R_C and how the trapping impacts R_C in particular. The interfacial charge-trap states in these systems could have originated from microstructural defects at the OSC surface/air interface.^[69] Overall, the reason for the reduced interfacial trap states in the AF2400 devices compared to the Cytop devices may be as simple as the AF2400 dielectric layer following the contour of the OSC more accurately than Cytop, and hence leading to a stronger coupling between the gate field and the channel layer. The POM images of the C₈-BTBT:C₁₆IDT-BT blend films in **Figure S7** show a number of bright spots, which were highlighted as tall aggregates in our earlier work.^[40] These defects likely contribute heavily to the charge trapping at the OSC/dielectric interface, and AF2400 is possibly better suited to making contact with the difficult topography of the aggregates than Cytop.

We used atomic force microscopy (AFM) to study the influence of the layer morphology on device operation. **Figure S8** shows topography images of the AF2400 and Cytop deposited onto the C₈-BTBT:C₁₆IDT-BT blend with the same optimized conditions used to produce the

best-performing OTFTs. The images show that the AF2400 has a much more intricate contour than the Cytop, which is significantly smoother. This may suggest that the AF2400 is highly conformal and capable of filling the various irregularities associated with the topography of the critical OSC/dielectric interface, when compared to the Cytop. Finally, a further possible contributor to the significant differences seen in the bias stability between the two dielectrics may be the large difference in their glass transition temperatures (T_g) of $\approx 110^\circ\text{C}$ for Cytop and $\approx 240^\circ\text{C}$ for AF2400.^[70] To this end, one can assume that the different T_g exerts a noticeable influence on the thermal conductivity of the dielectric which during device operation can affect the temperature of the semiconducting channel. However, analysis of such thermal effects is beyond the scope of this work.

3. Conclusions

We investigated the understudied relationship between the OSC/dielectric interface and the contact resistance, R_C , in hole mobility blend OTFTs. To do so, we compared two chemically similar fluoropolymer dielectrics, Cytop and AF2400, which are considered universal (compatible with hole and electron transporting OSCs) low-trapping materials, at best-performing conditions for C₈-BTBT:C₁₆IDT-BT OTFTs with a top-gate bottom-contact architecture. The devices were identical in all respects except for the material and thickness of the dielectric layers, allowing us to probe the impact of the OSC/dielectric interface on R_C without interference from other factors e.g., the potential energy barrier and charge trapping at the injecting metal/OSC interface as well as the OSC morphology. By calculating R_C at comparable electric flux densities, we found R_C to be worse in Cytop than AF2400-based transistors. To explore the idea that this difference in R_C was due to charge trapping at the OSC/dielectric interface, we compared the performances of the Cytop and AF2400 devices using bias-stress tests and density of states analyses. The results suggest that the

main difference between the two dielectric is more pronounced charge trapping at the Cytop/OSC interface than at the AF2400/OSC interface, further supporting the idea that accumulated charge trapping at the channel interface is related to the magnitude of R_C in OTFTs. Overall we highlight that, although R_C is in practice largely treated as a single figure, the charge injection process is complex and influenced by a number of factors, and the dielectric is one of the important considerations in the quest to resolve the infamous issue of R_C in OTFTs.

4. Experimental Section

Thin-film transistor fabrication and measurements: Top-gate bottom-contact transistors were fabricated on Borofloat glass slides. The source and drain electrodes were made from 40 nm thick gold by thermally evaporating through a shadow mask. The source and drain electrodes were then treated with pentafluorothiophenol (PFBT) to preferentially adjust the work function. The C₈-BTBT:C₁₆IDT-BT blend was made as described in previously reported in the literature,^{[16] [40]} at a total weight composition of 25% C₈-BTBT and 75% C₁₆IDT-BT, in a solvent blend comprised of 50% tetralin and 50% chlorobenzene. The solution was prepared at a concentration of 10 mg/ml. The solution was stirred overnight and heated at 60°C for 30 minutes prior to spin-coating. The thin-film was formed by spin-coating in two steps: (a) 500 rpm for 10 seconds and (2) 1500 rpm for 30 seconds. Immediately after spin coating, the film was annealed at 120°C for 2 minutes. The dielectric layer, made using either Cytop or poly[4,5-difluoro-2,2-bis(trifluoromethyl)-1,3-dioxole-*co*-tetrafluoroethylene] (Teflon™ AF2400), was deposited immediately after the C₈-BTBT:C₁₆IDT-BT blend. The Cytop was purchased from AGC Chemicals and the AF2400 was purchased from Sigma Aldrich. The AF2400 was diluted to a 25 mg/ml solution in FluorinertRTM FC-40, which was also purchased from Sigma Aldrich, and the solution was rolled and heated at 60°C for 5 days until it was fully

dissolved. The Cytop was spin coated at 2000 rpm for 60 seconds to produce a ≈ 1100 nm film, and the AF2400 was spin coated at 500 rpm for 20 seconds followed by 700 rpm for 30 seconds to produce a ≈ 300 nm film. Both dielectric layers were annealed at 50°C for 1 hour, and then a 50 nm aluminum gate electrode was thermally evaporated as the final step. After fabrication, a KEYSIGHT B2912A Precision Source/Measure Unit was used to analyse the thin-film transistors, to gather their current-voltage characteristics for device analysis, contact resistance and density of states calculations, as well as run bias-stress stability measurements.

Capacitance-frequency measurements: The capacitance values of the polymer dielectrics were determined in metal-insulator-metal structures sandwiching the respective polymer between an ITO bottom electrode and an Aluminium top electrode. The active area of each cell was 0.1 cm². Measurements were carried out inside a nitrogen filled glove box using a Solartron SI 1260 impedance analyser. Readings were taken within a frequency range of 100 Hz to 100 kHz, using a DC and AC level of 0 and 0.02 V, respectively.

Polarised optical microscopy: Polarised optical microscope images of the C₈-BTBT:C₁₆IDT-BT blend thin-films were taken using a Zeiss AX10 optical microscope.

Atomic force microscopy: Topographical information, regarding how the dielectric surfaces follow the contour of the semiconducting blend, was gathered using an Agilent 5500 atomic force microscope operating in tapping mode. Subsequent data analysis was undertaken using Gwyddion 2.39.

Supporting Information

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

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Figures

Figure 1. (a) Top-gate bottom-contact thin-film transistor architecture of the small-molecule/polymer blend OTFTs. (b) The chemical structure of pentafluorothiophenol (PFBT) used to modify the workfunction of the source and drain electrodes. Chemical structures of (c) the small-molecule 2,7-dioctyl[1]benzothieno[3,2-b][1]benzothiophene (C_8 -BTBT) and (d) the polymer indacenodithiophene - benzothiadiazole (C_{16} IDT-BT), which were the components of the semiconducting blend. (e) and (f) show the chemical structures of the fluoropolymer dielectric materials Cytop and AF2400, respectively.

Figure 2. Representative transistor current-voltage characteristics and contact resistance data for C₈-BTBT:C₁₆IDT-BT blend OTFTs, made with Cytop and AF2400 dielectric layers. (a) and (c) are plots of the transfer characteristics and V_G -dependent mobility, respectively, for the Cytop devices. (b) and (d) are plots of the transfer characteristics and V_G -dependent mobility, respectively, for the AF2400 devices. Both representative devices have channel lengths 50 μ m and width 1000 μ m. (e) Plot showing contact resistance dependency on electric flux density for C₈-BTBT:C₁₆IDT-BT blend OTFTs made with Cytop and AF2400. Dashed lines are pseudolinear fits given for eye guidance.

Figure 3. Comparison of 12 hour (12 h) bias-stress data for C₈-BTBT:C₁₆IDTBT blend OTFTs made with Cytop and AF2400. (a) Cytop and (b) AF2400 OTFT transfer curves before and after stressing, with plot of μ_{SAT} vs. V_G before and after stressing. OTFTs have a channel length of 80 μm and a width of 1000 μm . (c) Threshold voltage shift data showing how the threshold voltage has changed over time. The τ and β parameters were extrapolated from the stretched exponential fitting, which is shown by the solid lines.

Figure 4. Density of states (DOS) analysis calculated for C₈-BTBT:C₁₆IDT-BT blend devices made with both Cytop and AF400 dielectric layers, before (initial) and after 12 hour (12 h) of bias-stressing. The thermal equilibrium Fermi energy level (E_F) is the origin of the x-axis and corresponds to the OTFT flat-band voltage.

Table 1. Summary of key parameters for Cytop and AF2400 dielectric-based C₈-BTBT:C₁₆IDT-BT blend OTFTs.

Dielectric	Transistor parameters						
	C_i (F cm ⁻²)	V_{on} (V)	V_T (V)	$ V_T - V_{on} $ (V)	SS (V/dec.)	N_{tr} (cm ⁻²)	D_{tr} (cm ⁻² eV ⁻¹)
AF2400	5×10^{-9}	-2.5	-9.0	6.5	2.4	2.0×10^{11}	1.3×10^{12}
Cytop	1.7×10^{-9}	-6.0	-27.6	21.6	4.9	2.3×10^{11}	8.7×10^{11}

The type of gate dielectric in organic transistors is found to influence the contact resistance and bias stability of the devices. Both parameters are shown to vary between two different fluoropolymer dielectric materials namely Cytop and AF2400. Charge trapping at the interface between the dielectric and the organic semiconductor is argued to be the primary cause for this observation.

Impact of Gate Dielectric on Contact Resistance and Bias-Stress Stability of High Mobility Organic Transistors

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Supporting Information

Impact of the Gate Dielectric on Contact Resistance and Bias-Stress Stability of High Mobility Organic Transistors

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Figure S1. Capacitance-voltage measurements for the two dielectric layers, Cytop and AF2400.

Figure S2. Representative transfer and output curves for top-gate bottom-contact C₈-BTBT:C₁₆IDT-BT blend OTFTs made with Cytop and AF2400. (a) and (b) are the transfer and output curves, respectively, for the Cytop devices. (c) and (d) are the transfer and output curves, respectively, for the AF2400 devices. Both devices have channel length 50 μm and width 1000 μm .

Figure S3. Plot comparing contact resistance data for C₈-BTBT:C₁₆IDT-BT blend OTFTs made with Cytop and AF2400 at optimized processing conditions.

Figure S4. Transmission line method fits for C₈-BTBT:C₁₆IDT-BT transistors with (a) 1100 nm Cytop and (b) 300 nm AF2400. The fits were used to plot the contact resistance vs. gate voltage data shown in **Figure S3**.

Figure S5. Contact resistance vs. gate voltage data for C_8 -BTBT: C_{16} IDT-BT transistors made with varying thicknesses of Cytop as the dielectric layer: (a) 420 nm, (b) 445 nm, (c) 520 nm and (d) 585 nm. A representative transmission line method fit is shown in (e).

Figure S6. Contact resistance vs. gate voltage data for C_8 -BTBT: C_{16} IDT-BT transistors made with varying thicknesses of AF2400 as the dielectric layer: (a) 300 nm, (b) 380 nm, (c) 405 nm and (d) 680 nm. A representative transmission line method fit is shown in (e).

Figure S7. Polarised optical microscope images demonstrating that the morphology of the C₈-BTB:C₁₆IDT-BT organic blend semiconductor is the same in both the (a) Cytos and (b) AF2400 OTFTs.

Figure S8. Atomic force microscopy (AFM) images comparing the topography of (a) AF2400 and (b) Cytos, deposited on the surface of the C₈-BTBT:C₁₆IDTBT blend.

Table S1. Summary of the electric flux densities and corresponding contact resistance values, measured from Cytop and AF2400 dielectric layers of different thicknesses.

Flux density (Ccm ⁻²)	Cytop				AF2400			
	Thickness (nm)	C _i (nF/cm ²)	V _G (V)	R _C (Ωm)	Thickness (nm)	C _i (nF/cm ²)	V _G (V)	R _C (Ωm)
-3.0×10 ⁻⁷	585	3.25	-92	176	300	5.00	-60	25
-2.6×10 ⁻⁷	585	3.25	-79	111	405	3.69	-70	15
-2.2×10 ⁻⁷	520	3.65	-60	373	405	3.69	-60	40
-2.0×10 ⁻⁷	420	4.52	-45	1926	380	3.94	-50	163
-1.8×10 ⁻⁷	445	4.27	-42	656	405	3.69	-49	65
-1.6×10 ⁻⁷	420	4.52	-35	7119	380	3.94	-40	80
-1.5×10 ⁻⁷	445	4.27	-35	2447	405	3.69	-40	116
-1.3×10 ⁻⁷	445	4.27	-31	2740	680	2.20	-60	148
-1.1×10 ⁻⁷	520	3.65	-30	6345	680	2.20	-50	280
-1.0×10 ⁻⁷	420	4.52	-23	141703	300	5.00	-20	267
-3.7×10 ⁻⁷	520	3.65	-10	345275	405	3.69	-10	1444

To consider the contact resistance in the two dielectric systems fairly, both systems were compared at the same electric flux densities, to take into account the different electric fields and subsequent carrier densities experienced. To access a broader range of flux densities for the comparative data set, C₈-BTBT:C₁₆IDT-BT transistors were fabricated with both AF2400 and Cytop at varying dielectric layer thickness. The AF2400 dielectric layer was processed in one-step spin-coating speeds 1000 rpm (60s), 750 rpm (60s), 750 rpm (30s), and 500 rpm (30s) to give thicknesses 300, 380, 405 and 680 nm with areal capacitance values of 5.00, 3.94, 3.69 and 2.20, respectively. We note here that a new AF2400 solution made at a different concentration was used for these measurements, therefore the spin-speed is different to that detailed in the *Experimental Section*. The Cytop dielectric layer was processed in one-step spin-coating speeds 8000 rpm (60s), 7000 rpm (60s), 5000 (60s) and 4000 (60s) to give thicknesses 420, 445, 520 and 585 nm with areal capacitance values of 4.52, 4.27, 3.65 and 3.25 nF/cm², respectively. **Table S1** summarises the electric flux density values, calculated from the areal capacitance and measured gate voltages (*Equation 4*), and the corresponding R_C values in Ωm, as extracted from the R_C vs V_G plots shown in **Figure S5** and **Figure S6**. The results are shown in **Figure 2e**; using this approach allows R_C to be directly compared under the same carrier concentrations, as the OTFTs are based on the exact same architecture.