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Supplementary Materials for

Flexible low-voltage high-frequency organic thin-film transistors

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This PDF file includes:

Section S1 Figs. S1 to S7 Table S1 References

S1: Contact resistance, carrier mobility and transit frequency

The contact resistance (R_C) in organic TFTs is typically evaluated in the linear regime of transistor operation using the transmission line method (TLM; see Figs. 2 and S4) (*41*). In TLM analyses, as well as for most other methods for determining $R_C(42)$, a requirement (and indeed a drawback) is that it is only valid for the linear regime of operation using vanishingly-small *V_{DS}*, since a key assumption of TLM is that R_C is Ohmic and that the channel resistance is uniform over the entire channel region. This latter point by itself precludes any analysis of *R^C* in the saturation regime using TLM because of the pinch-off of the channel that occurs when *VDS* is approximately equivalent to the overdrive voltage $(V_{GS} - V_{th})$. Experimental investigations have shown that R_C in organic TFTs can vary significantly with V_{DS} (32, 43). Especially for TFTs with small channel lengths (L) , R_C may be reduced by V_{DS} due to effects such as image-force lowering (IFL) (*33*) or by drain-induced barrier lowering (DIBL) (*35*) of the injection barrier at the interface between the source contact and the semiconductor. Enhancements of the carrier mobility in the vicinity of the contacts through a Poole-Frenkel-like dependence of the mobility on the applied electric field may also lead to lower R_C , in part due to the nonlinear effects on both charge transport (*44*) and charge injection (*45*). For these reasons it is beneficial to be able to quantify the dependence of R_C on V_{DS} .

Here, we estimate R_C and the intrinsic channel mobility (μ_0) in the saturation regime ($V_{DS} \leq$ V_{GS} - V_{th} for p-channel TFTs) by fitting the transit frequency (f_T) extracted from *S*-parameter measurements as a function of channel length (*L*). Similar to TLM, we use μ_0 and the widthnormalized contact resistance (R_CW) as fitting parameters. This approach has to our knowledge never been explicitly implemented, though comparisons of calculated *f^T* to experimental results using *R^C* determined from TLM have been reported (*16*, *36*). In both approaches, the error in the extracted values is assessed simply as the calculated standard error from the fits. In the TLM, the width-normalized total source-to-drain resistance of the TFT (*RW*) is fit with a linear function with respect to *L*. In our method, the f_T data as a function of *L* is fit using the equation derived in the following.

The dependence of the effective mobility (μ_{eff}) and the transit frequency (f_T) on R_C can be illustrated with the following two equations for TFTs operated in the saturation regime (*2*, *46*):

$$
\mu_{eff} = \mu_0 \left[1 - \left(\frac{\mu_0 C_{diel} R_C W(V_{GS} - V_{th})}{L + \mu_0 C_{diel} R_C W(V_{GS} - V_{th})} \right)^2 \right]
$$
(1)

$$
f_T = \frac{\mu_{eff}(V_{GS} - V_{th})}{2\pi L \left(L_{ov, total} + \frac{2}{3}L\right)}\tag{2}
$$

where *C*_{diel} is the gate-dielectric capacitance per unit area and *L*_{ov,total} is the total gate-to-contact overlap length, which is simply the sum of the gate-to-source (*Lov,GS*) and gate-to-drain (*Lov, GD*) overlap lengths. In principle, the term R_C encompasses contributions from both the source and drain contacts. Equation 1 can be simplified to (*36*)

$$
\mu_{eff} = \frac{\mu_0}{1 + \frac{1}{2L}\mu_0 C_{diel} R_C W (V_{GS} - V_{th})}
$$
(3)

Combining Equations 2 and 3, we then obtain an expression for f_T that includes the influence of the contact resistance:

$$
f_T = \frac{\mu_0 (V_{GS} - V_{th})}{2\pi \left(L + \frac{1}{2}\mu_0 C_{diel} R_C W (V_{GS} - V_{th}) \right) \left(L_{ov,total} + \frac{2}{3}L \right)} \tag{4}
$$

For illustrative purposes, several curves of the transit frequency as a function of the channel length calculated using Equation 4 and assuming different values for *RCW* are shown in Fig. S7.

Supplementary Figures

Fig. S1| Device fabrication process and materials characterization. (A) Schematic process flow for the fabrication of bottom-gate bottom-contact (inverted coplanar) organic TFTs. All metal and semiconductor layers are deposited by thermal evaporation or sublimation in vacuum and patterned using high-resolution silicon stencil masks. **(B)** Infrared reflection absorption spectroscopy (IRRAS) analysis of bulk pentafluorobenzenethiol (PFBT, black) and of a chemisorbed monolayer of PFBT on a gold surface (red). **(C)** AFM height scan of a thin film of the organic semiconductor DPh-DNTT deposited onto a hybrid AlO_x/SAM gate dielectric on a flexible PEN substrate.

Fig. S2| Static transistor characteristics and uniformity. (A) SEM micrograph of an individual DPh-DNTT TFT. **(B)** Measured transfer characteristics of 10 nominally identical TFTs having a channel length (*L*) of 1.5 µm, a total gate-to-contact overlap (*Lov,total*) of 60 µm and a channel width (*W*) of 7.5 μ m, with statistics for the effective carrier mobility (μ_{eff}), threshold voltage (*Vth*) and subthreshold swing (*SS*). **(C)** Transfer characteristics of an individual TFT measured at a drain-source voltage (V_{DS}) of -3 V. The dotted blue line is a guide to the eye, indicating the ideal quadratic dependence of the drain current on the gate-overdrive voltage (*VGS* $-V_{th}$) in the saturation regime. **(D)** Output characteristics for gate-source voltages (V_{GS}) from 0 to -3 V in steps of 0.5 V.

Fig. S3| Static characteristics of TFTs based on DPh-DNTT (top row) and C10-DNTT (bottom row). Both TFTs have a channel length (*L*) of 8 µm, a total gate-to-contact overlap (*Lov,total*) of 4 µm and a channel width (*W*) of 200 µm. From left to right: Transfer characteristics, effective carrier mobility (μ_{eff}) plotted as a function of the gate-source voltage, and output characteristics for gate-source voltages (V_{GS}) from 0 to -3 V in steps of 0.5 V. (Note that these TFTs were fabricated separately from the TFTs shown in Fig. S2.)

Fig. S4| Transistors for TLM analysis. (A) Transfer characteristics of DPh-DNTT TFTs with channel lengths (L) ranging from 1 to 10.5 μ m and a channel width (W) of 50 μ m, measured with a drain-source voltage (V_{DS}) of -0.1 V. **(B)** Effective carrier mobility (μ_{eff}) in the linear regime determined for each TFT as a function of the channel length (*L*). The line is a fit to the data using the equation $\mu_{eff} = \mu_0 (1 + L_{I/2}/L)^{-1}$ where μ_0 is the intrinsic channel mobility and $L_{I/2}$ is the channel length at which $\mu_{\text{eff}} = \frac{1}{2} \mu_0$. (C) SEM micrographs of the channel region of the TFTs. (Note that these TFTs were fabricated separately from the TFTs shown in Fig. S2.)

Fig. S5| Transistors for *S***-parameter measurements. (A)** Transfer characteristics of DPh-DNTT TFTs with channel lengths (*L*) ranging from 0.7 to 10.5 µm, a total gate-to-contact overlap (*Lov,total*) of 10 µm and a channel width (*W*) of 100 µm, measured with a drain-source voltage (V_{DS}) of -3 V. **(B)** Output characteristics of each TFT for a gate-source voltage (V_{GS}) of -3 V. **(C)** Channel-width-normalized peak transconductance (*gm*) at a gate-source voltage (*VGS*) of -3 V as a function of the inverse of the channel length. **(D)** SEM micrographs of the channel region of the TFTs. (Note that these TFTs were fabricated separately from the TFTs shown in Fig. S2.)

Fig. S6| DPh-DNTT TFT showing a transit frequency of 21 MHz. The TFT has a channel length (*L*) of 0.6 µm, a total gate-to-contact overlap (*Lov,total*) of 10 µm and a channel width (*W*) of 100 µm. **(A)** Output characteristics for gate-source voltages (*VGS*) from 0 to -3 V in steps of 0.5 V. **(B)** SEM micrographs of the channel region of the TFT.

Fig. S7| Relation between channel length, contact resistance and transit frequency. The curves were calculated using Equation (4) for width-normalized contact resistances (R_C *W*) of 10, 50 and 100 Ωcm, gate-to-source and gate-to-drain overlaps (*Lov,GS, Lov,GD*) of 5 µm, an intrinsic channel mobility (μ_0) of 5 cm²/Vs and a gate-overdrive voltage ($|V_{GS}$ - $V_{DS}|$) of 2 V.

Table S1| Literature review of voltage-normalized transit frequencies reported for organic TFTs.

^a Small-signal currents directly measured to evaluate f_T .

 $\sum_{i=1}^{b} S$ -parameter measurement to evaluate f_T .

^c Pulsed-bias measurement circuit to evaluate f_T .

*In cases where the data were obtained from measurements on ring oscillators (RO), the equivalent frequency $f_{eq} = 1/(2\tau)$ is normalized to the supply voltage.

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