Supporting Information

Improving Electron Injection and Transport in Polymer Field-Effect Transistors with Guanidino-Functionalized Aromatic n-Dopants

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Atomic Force Microscopy of P(NDI2OD-T2) Films



Figure S1. Atomic force microscopy (AFM) images $(2.5 \times 2.5 \ \mu\text{m}^2, \text{ all scale bars 500 nm})$ of P(NDI2OD-T2) films corresponding to the untreated reference sample (a) and with ttmgb top layers that were evaporated (b) or spin-coated from toluene solution (c). AFM images of P(NDI2OD-T2) films spincoated on top of injection layers of ttmgb (d) and tdmegb (e), as well as blends of P(NDI2OD-T2) with 0.5 w% (f) and 1.0 w% (g) ttmgb.



Output Characteristics of P(NDI2OD-T2) FETs with Injection Layers

Figure S2. Output characteristics of P(NDI2OD-T2) FETs ($L = 40 \ \mu m$, $W = 5 \ mm$) with dopant injection layers corresponding to data presented in Fig. 2 of the main part and measured at $V_g = 0 - 10$ V in steps of 2.0 V for reference (a), with thin (b) and thick (c) ttmgb injection layers. Respective data for tdmegb layers (d-f). Insets show zoom-ins on the low drain voltage (linear) regime that indicates injection properties.



Output Characteristics of FETs with P(NDI2OD-T2)/ttmgb Blends

Figure S3. Output characteristics of FETs ($L=40 \ \mu m$, $W=5 \ mm$) with P(NDI2OD-T2)/ttmgb blends corresponding to data presented in Fig. 3 of the main part and measured at $V_g = 0 - 10 \ V$ in steps of 2.0 V for reference (a), with 0.1 w% (b) and 2.0 w% ttmgb (c) blend layers. Insets highlight the injection behavior of the corresponding FET.

Output Characteristics of P(NDI2OD-T2) FETs with ttmgb Top Layers



Figure S4. Output characteristics of P(NDI2OD-T2) FETs ($L=40 \mu m$, W=5 mm) with ttmgb top layers corresponding to data presented in **Fig. 4** of the main part and measured at $V_g = 0 - 10$ V in steps of 2.0 V for reference (a), with evaporated (b) and spin-coated (c) ttmgb top layers. Insets highlight the injection behavior of the corresponding FET.

Transport Characteristics of DPPT-BT FETs with Different Injection Layers



Figure S5. DPPT-BT FETs ($L = 42 \ \mu m$, $W = 1.5 \ mm$) with ttmgb injection layers. (a) Simplified device schematic and output characteristics of thick ttmgb injection layer (solid lines) and untreated reference sample (dotted lines). Transfer characteristics of FETs in linear (b) and saturation regime (c) for untreated reference sample and three dopant layer thicknesses for solution-processed ttmgb. (d) Corresponding electron mobility in linear (black squares) and saturation regime (red circles). Error bars represent standard deviation based on at least 6 measured FETs.

Atomic Force Microscopy of DPPT-BT Thin Films



Figure S6. Atomic force microscopy images $(2.5 \times 2.5 \ \mu m^2)$, all scale bars 500 nm) of DPPT-BT films representing the untreated reference sample (a), with ttmgb injection layer (b) and spin-coated (c) and evaporated (d) ttmgb top layers.



Output Characteristics of DPPT-BT FETs with ttmgb Top Layers

Figure S7. Output characteristics of DPPT-BT FETs ($L=40 \ \mu m$, $W=5 \ mm$) with ttmgb top layers corresponding to data presented in **Fig. 7** of main part and measured at $V_g = 0 - 10$ V in steps of 2.0 V for reference (a), with evaporated (b) and spin-coated (c) ttmgb top layers. Insets show zoomins on the low drain voltage (linear) regime that indicates injection properties.

Transfer Characteristics of DPPT-BT FETs Depending on Dopant Layer Thickness



Figure S8. DPPT-BT FETs ($L = 40 \ \mu m$, $W = 5 \ mm$) with ttmgb top layers. Simplified device schematic and transfer characteristics of untreated reference FET and FETs with different thicknesses of spin-coated and evaporated ttmgb layers in the linear (a) and saturation regime (b).

Gate Voltage-Dependent Electron Mobility Extracted from DPPT-BT FETs



Figure S9. Gate voltage-dependent charge carrier mobilities extracted from DPPT-BT FETs $(L = 40 \text{ }\mu\text{m}, W = 5 \text{ }\text{mm})$ for untreated reference FET (black) and FETs with thick ttmgb solution-processed top layer (red) in the linear (full squares) and saturation (empty circles) regime.

Experimental Parameters Spin-Coated ttmgb Layers

Polymer	Architecture	Layer label in manuscript	ttmgb concentration (g·L ⁻¹)	Spincoating speed (rpm)
P(NDI2OD-T2)	Injection Layer	Thin	1.4	4,000
P(NDI2OD-T2)	Injection Layer	Thick	2.8	4,000
P(NDI2OD-T2)	Top Layer	n/a	0.5	1,000
DPPT-BT	Injection Layer	Thin	1.4	4,000
DPPT-BT	Injection Layer	Medium	2.8	4,000
DPPT-BT	Injection Layer	Thick	5.6	4,000
DPPT-BT	Top Layer	Thin	1.0	1,000
DPPT-BT	Top Layer	Medium	2.0	1,000
DPPT-BT	Top Layer	Thick	4.0	1,000

 Table S1. Parameters for spin-coated ttmgb layers.

Experimental Parameters Vacuum-Deposited Dopant Layers

Table S2. Parameters for evaporated dopant layers.

Polymer	Dopant	Architecture	Layer Label in Manuscript	Layer Thickness (nm)*
P(NDI2OD-T2)	tdmegb	Injection Layer	Thin	0.8
P(NDI2OD-T2)	tdmegb	Injection Layer	Thick	2.0
P(NDI2OD-T2)	ttmgb	Top Layer	n/a	1.1
DPPT-BT	ttmgb	Top Layer	Thin	2.0
DPPT-BT	ttmgb	Top Layer	Thick	4.0

*amorphous/polycrystalline on glass, not homogeneous on metal electrode/on top of polymer layer