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Supporting Information

Stability of Organic Thin-Film Transistors based on Ultrathin Films of Dinaphtho[2,3-*b*:2',3'*f*]thieno[3,2-*b*]thiophene (DNTT)

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Figure S1: AFM image showing the morphology of a nominally 2.5-nm-thick DNTT film deposited onto a hybrid AlO_x/n -tetradecylphosphonic acid SAM dielectric, and height profile indicating a characteristic step height of 1.6 nm, confirming that the DNTT molecules stand approximately upright on the substrate surface.



- Figure S2:Evolution of the morphology of nominally 2.5-nm-thick DNTT films deposited onto
(a) bare AlOx (without SAM);
(b) AlOx covered with a SAM of *n*-hexylphosphonic acid ($C_6H_{13}PA$);
(c) AlOx covered with a SAM of *n*-octadecylphosphonic acid ($C_{18}H_{37}PA$).
 - The AFM images were recorded within 15 minutes after the completion of the DNTT deposition and again after 12 hours.



Figure S3: Specular X-ray diffractogram of a nominally 10-nm-thick DNTT film deposited onto a hybrid AlO_x/n -tetradecylphosphonic acid SAM dielectric together with a calculated powder spectrum. The inset schematically depicts the molecular arrangement.



Figure S4: Output and transfer curves and charge-carrier mobility as a function of gate-source voltage of

(a) a TFT with a nominally 2.5-nm thick DNTT film;

(b) a TFT with a nominally 25-nm thick DNTT film.

The transfer curves were recorded within 15 minutes after the completion of the DNTT deposition.



Figure S5: Morphology of a nominally 25-nm-thick thick DNTT film deposited onto AlO_x covered with a SAM of *n*-tetradecylphosphonic acid ($C_{14}H_{29}PA$). The AFM images were recorded within 15 minutes after the completion of the DNTT deposition and again after 12 hours.



Figure S6: Evolution of the morphology of

(a) nominally 2.5-nm-thick DNTT without encapsulation;

(b) nominally 2.5-nm-thick PTFE on nominally 2.5-nm-thick DNTT;

(c) nominally 2.5-nm-thick C_{60} on nominally 2.5-nm-thick DNTT;

(d) nominally 2.5-nm-thick TiOPc on nominally 2.5-nm-thick DNTT.

The AFM images were recorded within 15 minutes after the completion of the deposition of the encapsulation film and again after 72 hours.



Figure S7: Evolution of the morphology of a nominally 2.5-nm-thick PTFE film deposited onto an AlO_x/n -tetradecylphosphonic acid SAM dielectric. The AFM images were recorded within 15 minutes after the completion of the PTFE deposition and again after 12 hours and after 36 hours.



Figure S8: Evolution of the morphology of a nominally 10-nm-thick PTFE film deposited onto a nominally 2.5-nm-thick DNTT film. The AFM images were recorded within 15 minutes after the completion of the PTFE deposition and again after 72 hours.



Figure S9: (a) Evolution of the subthreshold swing of TFTs based on vacuum-deposited DNTT films with nominal thicknesses of 2.5 and 25 nm with and without encapsulation with PTFE, C₆₀ or TiOPc with various nominal thicknesses.
(b) Transfer curves of TFTs based on vacuum-deposited DNTT films with nominal thicknesses of 2.5 and 25 nm with and without encapsulation, recorded within 30 minutes after the completion of the last deposition.

Table S1:	Summary of the root-mean-square surface roughness (R _{RMS}) values extracted from
	each AFM image.

Figure	Image	R _{RMS} [nm]
Figure 1	1.0 nm DNTT	1.86
	1.5 nm DNTT	1.29
	2.0 nm DNTT	0.75
	2.5 nm DNTT	0.69
Figure 2	15 minutes	0.69
	1 hour	0.94
	3 hours	1.41
	6 hours	1.83
	9 hours	2.34
	12 hours	2.61
Figure 4	a) After deposition	1.60
	b) Stored @ 193 K	1.69
	c) Stored @ 300 K	17.09
Figure S2	a) 15 minutes	1.37
	a) 12 hours	2.86
	b) 15 minutes	0.96
	b) 12 hours	3.15
	c) 15 minutes	1.93
	c) 12 hours	2.38
Figure S5	15 minutes	4.09
	12 hours	4.12
Figure S6	a) 15 minutes	1.60
	a) 72 hours	6.47
	b) 15 minutes	3.37
	b) 72 hours	5.29
	c) 15 minutes	3.14
	c) 72 hours	4.44
	d) 15 minutes	1.57
	d) 72 hours	1.61
Figure S7	15 minutes	2.33
	12 hours	3.23
	36 hours	3.79