**Supporting Information for**

**All-brush-painted top-gate organic thin-film transistors**

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Fig. S1 Schematic of FTS patterning process by using a-c) PMMA film and d-e) metal mask as the pattern model. a) Deposition of PMMA layer on FTS modified substrate, b) patterning PMMA followed by the O_2 plasma treatment and c) SiO_2/Si substrate with patterned FTS. d) Covering metal mask on the FTS modified substrate followed by the UVO treatment and e) glass substrate with patterned FTS.
Fig. S2 Chemical structure of poly(3-hexylthiophene) (P3HT) and poly(2,5-bis(3-alkylthiophen-2-yl)thieno[3,2-b]thiophene) (PBT TT).  

Fig. S3 The XRD spectra of thin film based on P3HT and PBT TT, prepared by spin coating and brush painting at different temperatures and followed by annealing treatment at 100 °C for P3HT and 170 °C for PBT TT.
Fig. S4 Transfer characteristics of OTFTs with brush painted a) P3HT dissolved in chloroform, b) in chlorobenzene and c) PBTTT dissolved in dichlorobenzene utilizing pre-patterned Au as source-drain electrodes, spin-coated PMMA as dielectric film, and evaporated silver as gate electrode. (a: W=2800 µm, L=30 µm; b: W=2800 µm, L=40 µm; c: W=2800 µm, L=10 µm)

Fig. S5 Leak current test of two-terminal device with structure of ITO/PMMA/Ag.
Fig. S6 Transfer and output characteristics of P3HT based all-brush-painted devices at different substrate temperatures of a-b) 70 °C, c-d) 100 °C, and e-f) 170 °C. (W=4200 μm, L=100 μm)
Fig. S7 Transfer and output characteristics of PBTTT based all-brush-painted devices at different substrate temperatures of a-b) 70 °C, c-d) 100 °C, and e-f) 170 °C.(a,b: W=2800 µm, L=80 µm; c,d: W=1800 µm, L=100 µm; e,f: W=2000 µm, L=60 µm)
Fig. S8 Statistical histograms of field-effect mobility for P3HT based OTFTs brush painted at a) 70 °C, b) 100 °C, c) 130 °C and d) 170 °C.

Fig. S9 Statistical histograms of field-effect mobility for PBTTT based OTFTs brush painted at a) 70 °C, b) 100 °C, c) 130 °C and d) 170 °C.
Fig. S10 a) Transfer characteristics of P3HT based devices with all functional layers deposited by brush painting in ambient conditions. b) Transfer curve of a P3HT based device before (black) and after (red) storage for 9 days in air. (a: W=3000 µm, L=140 µm)