Electronic Supplementary Information

Achieving high field-effect mobility in CuSCN thin-film transistors with thiocyanate-functionalized polymers as fluorine-free dielectrics

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Table S1. Compositions used for synthesizing thiocyanate-containing polymers, each with number and weight average molecular weight (M_n and M_w) and molar weight distribution (MWD = M_w/M_n).

Entry	PVBC		KSCN		M_n	M_w	MWD
	mg	mmol	mg	mmol	g mol^{-1}	g mol^{-1}	
PVBC	-	-	-	-	35,000	70,100	1.84
P(VBC$_{0.80}$-*co*-VBT$_{0.20}$)	152.6	1.00	19.4	0.20	28,700	46,200	1.61
P(VBC$_{0.49}$-*co*-VBT$_{0.51}$)	152.6	1.00	48.6	0.50	28,300	48,900	1.73
PVBT	152.6	1.00	113.7	1.17	29,100	55,300	1.90

[a] Calculated from ^{1}H-NMR spectroscopy.

Preparation of polymer blend

The homopolymer blend was prepared from the stock solution of each homopolymer (PVBC and PVBT) at 60 mg mL^{-1}. Then, to obtain a precise mixture with Cl:SCN = 1:1 (mol:mol), the homopolymer solutions were mixed and yield a final solution of 935 μL by the calculation listed in **Table S2**.

Table S2. Calculations for preparing the blend.

Parameter	PVBC	PVBT
Molecular mass of monomer unit (g mol^{-1})	152.63	175.25
Desired molar fraction	0.50	0.50
Corresponding measured weight fraction	0.47	0.53
Volume used for preparing a mixture (μL)	435	500

Standard TFT parameters

In the main text, TFT parameters were converted to capacitance-weighted parameters due to the difference in the dielectric constant for each polymer. **Table S3** below lists the standard TFT parameters including the hole mobility in saturation regime ($\mu_{h,sat}$), threshold voltage (V_{th}), subthreshold swing (S_{th}) and on-off current ratio (I_{on}/I_{off}).

Table S3. Summary of TFT parameters derived from forward scans of at least 8 devices for each polymer dielectric.

Polymer dielectric	$\mu_{h,sat}$ ($\times10^{-2}$ cm^2 V^{-1} s^{-1})			V_{th} (V)			S_{th} (V dec^{-1})			I_{on}/I_{off} ($\times10^3$)		
	Max	Avg	SD	|Min|	Avg	SD	Min	Avg	SD	Max	Avg	SD
P(VBC$_{0.80}$-*co*-VBT$_{0.20}$)	1.84	1.66	0.12	-18.9	-19.4	0.3	4.06	4.15	0.10	1.5	1.2	0.2
P(VBC$_{0.49}$-*co*-VBT$_{0.51}$)	1.73	1.53	0.13	-7.3	-7.6	0.2	2.18	2.50	0.18	2.2	1.8	0.2
PVBT	3.01	2.63	0.36	-1.9	-2.7	0.5	1.94	2.16	0.20	2.0	1.6	0.3

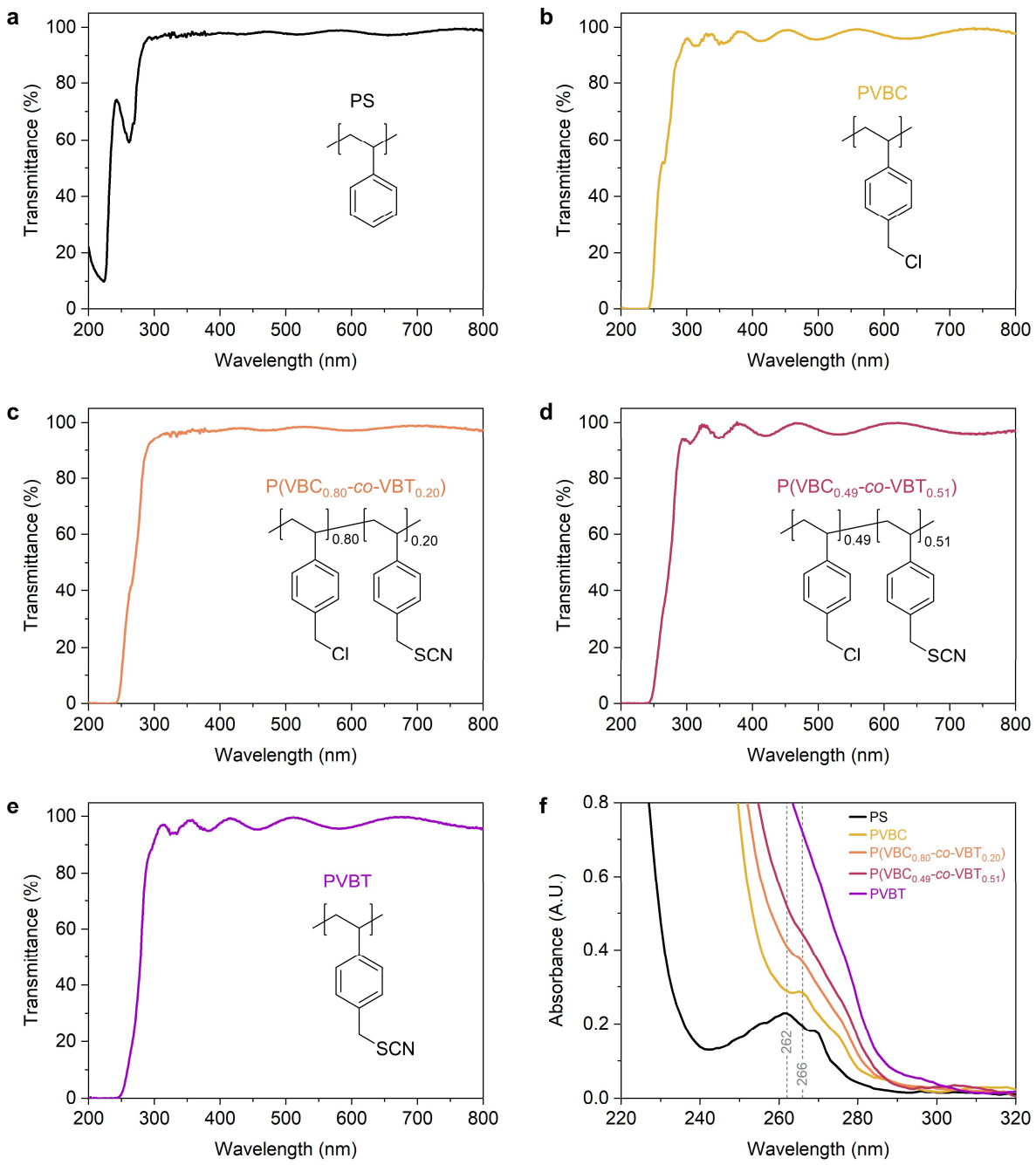

Figure S1. Optical transmittance of (a) PS, (b) PVBC, (c) P(VBC$_{0.80}$-*co*-VBT$_{0.20}$), (d) P(VBC$_{0.49}$-*co*-VBT$_{0.51}$), and (e) PVBT. (f) Comparison of spectra near the absorption edge.

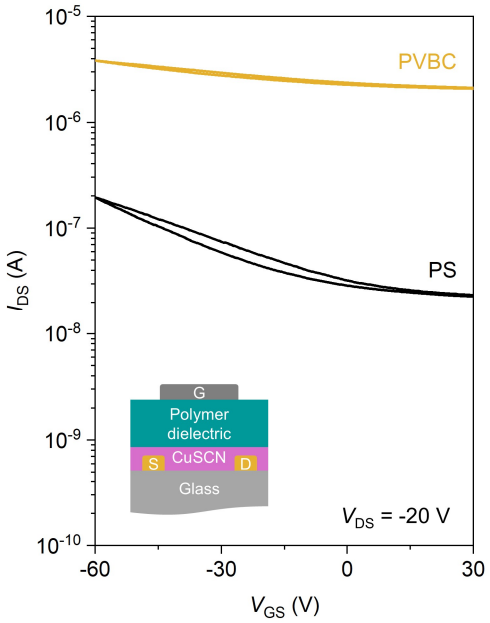

Figure S2. Transfer characteristics of CuSCN TFTs based on PS and PVBC dielectrics.

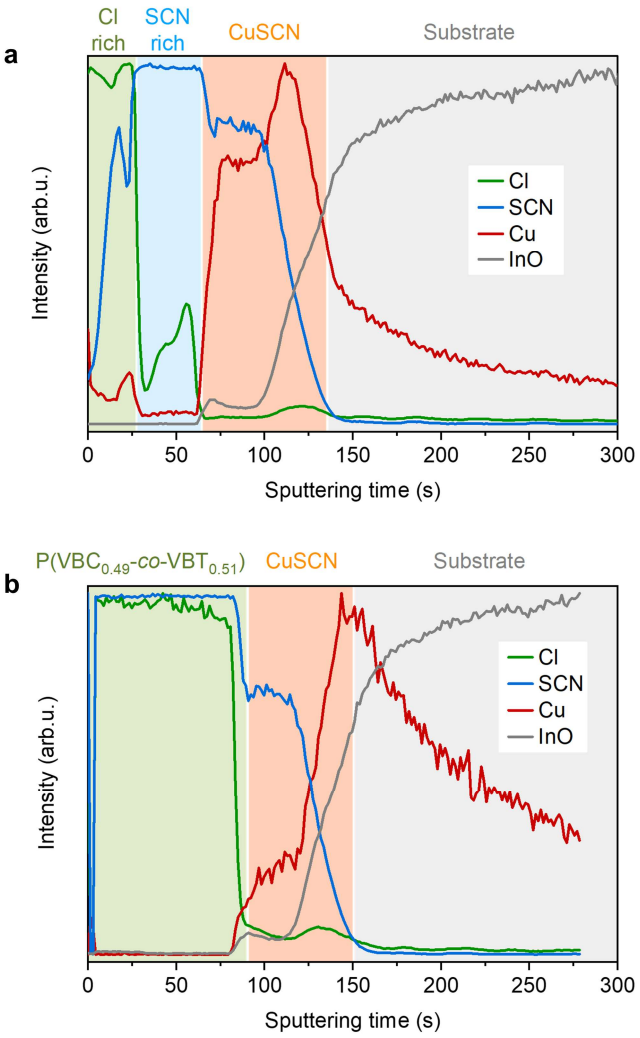

Figure S3. Elemental profiles measured by secondary-ion mass spectrometry (SIMS) of (a) a blend of homopolymers PVBC:PVBT at 0.47:0.53 by weight (equivalent to Cl:SCN of 1:1 molar ratio) and (b) the copolymer P($VBC_{0.49}$-*co*-$VBT_{0.51}$). Films were spin-coated from solutions with a concentration of 60 mg mL^{-1} in THF.

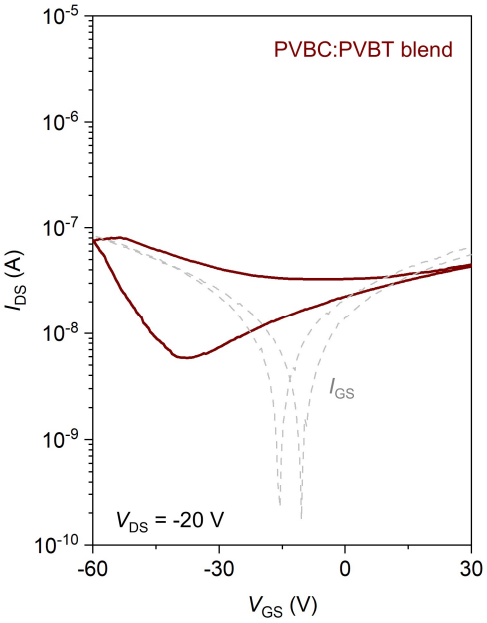

Figure S4. Transfer curve of a CuSCN TFT based on a homopolymer blend of PVBC:PVBT with a 0.47:0.53 weight ratio. The dielectric blend was spin-coated from a 60 mg mL^{-1} solution in THF.