

Electronic Supplementary Information

Furan-Flanked Diketopyrrolopyrrole-Based Chalcogenophene Copolymers with Siloxane Hybrid Side Chains for Organic Field-Effect Transistors

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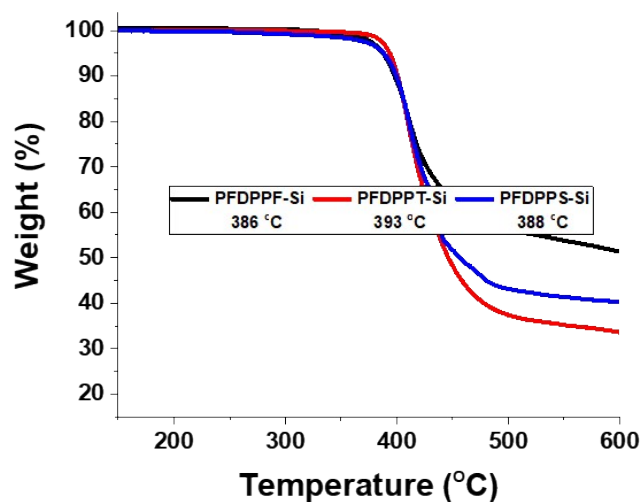


Fig. S1. TGA plots of **PFDPPX-Si** copolymers with a heating rate of 10 °C/min under a nitrogen atmosphere and an inset box indicating each temperature at 5% weight loss.

Table S1. Electrical properties of **PFDPPX-Si** from the DFT calculations.

Copolymer	E_{HOMO} (eV)	E_{LUMO} (eV)	E_{g} (eV)
PFDPPF-Si	-4.66	-2.84	1.82
PFDPPT-Si	-4.64	-2.87	1.77
PFDPPS-Si	-4.62	-2.88	1.74

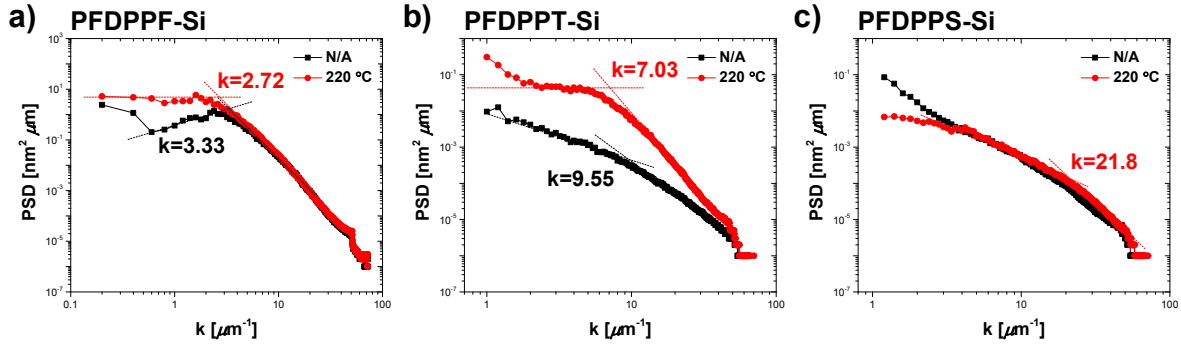


Fig. S2. PSD analysis of the AFM images presented in Fig. 3 for (a) **PFDPPF-Si**, (b) **PFDPPT-Si**, and (c) **PFDPPS-Si** films before (black line) and after (red line) annealing at 220 °C.

Table S2. PSD analysis parameters of as-cast and annealed **PFDPPX-Si** polymer films.

Conditions	As-cast (N/A)		Annealed at 220 °C	
	Spatial frequency, k [μm^{-1}]	Correlation length [nm]	Spatial frequency, k [μm^{-1}]	Correlation length [nm]
PFDPPF-Si	3.33	300	2.72	368
PFDPPT-Si	9.55	105	7.03	142
PFDPPS-Si	— ^a	— ^a	21.8	45.9

^a The spatial frequency and correlation length could not be extracted.

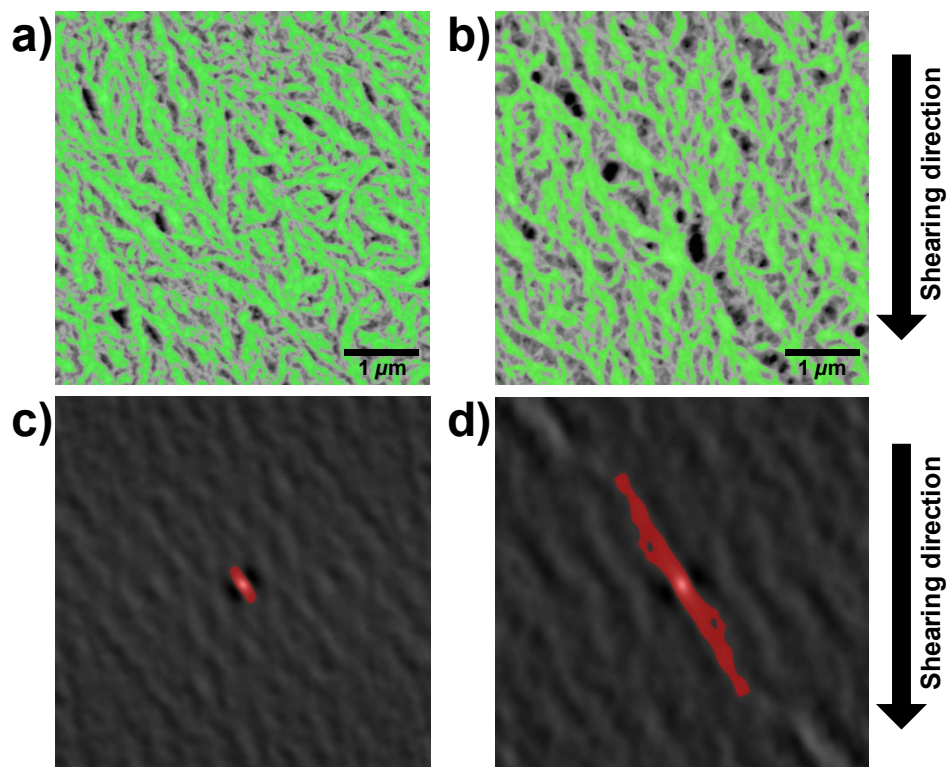


Fig. S3. The height thresholding of **PFDDPF-Si** films (a) before and (b) after thermal annealing at 220 °C. Shearing force was applied through the vertical direction to form polymer films. The grains marked by green were formed in the shearing direction. The two-dimensional autocorrelation of **PFDDPF-Si** films (c) before and (d) after thermal annealing at 220 °C with threshold 0.05. The film anisotropy (red spot) was evaluated to be aligned through shearing direction.

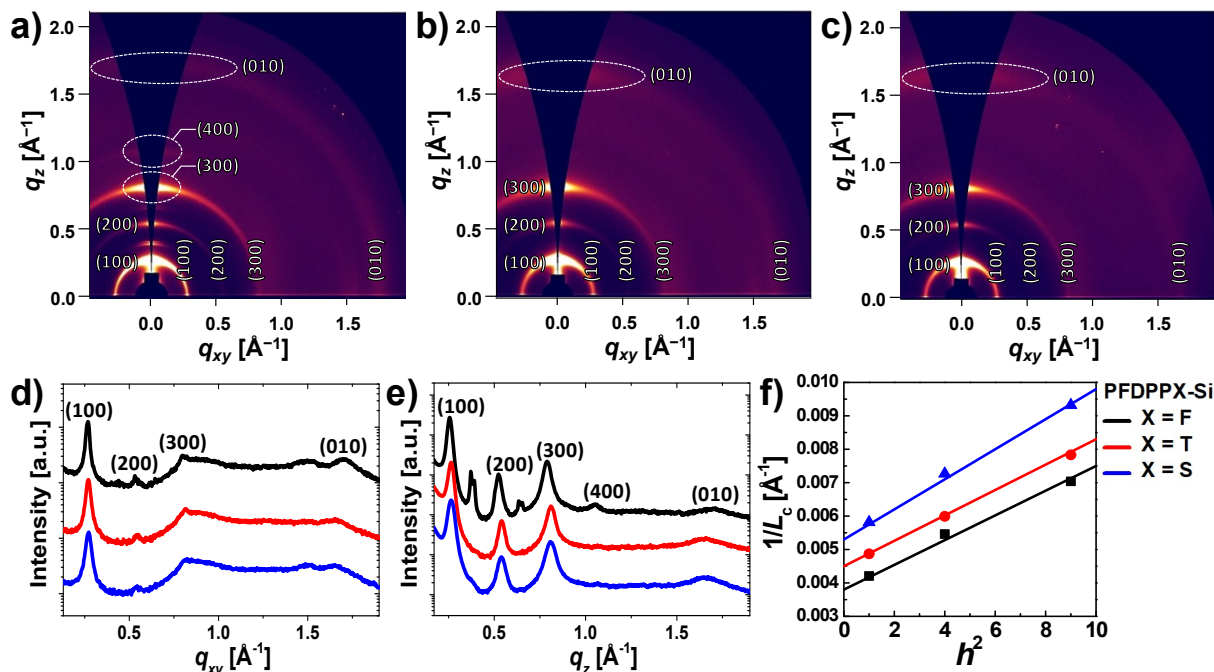


Fig. S4. 2D-GIXD images of as-cast a) **PFDPFF-Si**, b) **PFDPPT-Si**, and c) **PFDPSS-Si** films prepared by solution shearing method on OTS-treated SiO₂/Si substrates with shearing speed of 0.1 mm s⁻¹. The corresponding X-ray diffractogram profiles of d) in-plane and e) out-of-plane GIXD patterns. f) Williamson-Hall plots of the diffraction peaks obtained from out-of-plane direction; black lines for **PFDPFF-Si**, red lines for **PFDPPT-Si**, and blue lines for **PFDPSS-Si**.

Table S3. Crystallographic parameters of as-cast **PFDPXX-Si** polymer films.^a

Copolymer	Lamellar spacing					π - π stacking			
	$q_{z,(100)}$ [Å ⁻¹]	$d_{z,(100)}$ [Å]	$\text{FWHM}_{(100)}$ [Å ⁻¹]	$L_{z,(h00)}^b$ [Å]	g^c [%]	$q_{z,(010)}$ [Å ⁻¹]	$q_{xy,(010)}$ [Å]	$q_{xy,(010)}$ [Å ⁻¹]	$d_{xy,(010)}$ [Å]
PFDPFF-Si	0.256	24.6	0.0238	263.2	3.03	1.70	3.69	1.71	3.68
PFDPPT-Si	0.263	23.9	0.0276	222.2	3.03	1.66	3.79	1.65	3.82
PFDPSS-Si	0.264	23.8	0.0329	188.7	3.30	1.66	3.77	1.67	3.76

^a The polymer films were prepared by solution shearing with shearing speed of 0.1 mm s⁻¹ without further treatment, and their parameters were calculated from GIXD profiles. ^b True crystallite sizes were obtained from the intercept of the straight line with the ordinate of the Williamson-Hall plot.

^c Lattice disorder parameter is related to slope of the Williamson-Hall plot.

Table S4. Summary of the electrical characteristics of OFETs with **PFDPPX-Si** films under different conditions.

Conditions ^a		<i>p</i> -Channel				<i>n</i> -Channel		
Copolymer	Films	T _a [°C]	μ _{max} [cm ² V ⁻¹ s ⁻¹]	μ _{avg} ^b [cm ² V ⁻¹ s ⁻¹]	I _{on} /I _{off}	μ _{max} [cm ² V ⁻¹ s ⁻¹]	μ _{avg} [cm ² V ⁻¹ s ⁻¹]	I _{on} /I _{off}
PFDPPF-Si	Drop cast	N/A ^c	1.03	0.78 (±0.149) ^d	>10 ⁷	0.06	0.05 (±0.005)	>10 ³
		220	1.80	1.16 (±0.339)	>10 ⁶	0.10	0.07 (±0.020)	>10 ³
	Solution sheared	N/A	1.20	0.86 (±0.178)	>10 ⁵	0.12	0.08 (±0.024)	>10 ⁴
		220	2.48	1.80 (±0.473)	>10 ⁴	0.16	0.12 (±0.031)	>10 ³
PFDPPT-Si	Drop cast	N/A	0.84	0.69 (±0.122)	>10 ⁷	0.12	0.10 (±0.010)	>10 ³
		220	1.56	1.12 (±0.213)	>10 ⁷	0.18	0.13 (±0.030)	>10 ⁴
	Solution sheared	N/A	0.94	0.76 (±0.120)	>10 ⁶	0.14	0.12 (±0.016)	>10 ³
		220	1.99	1.43 (±0.204)	>10 ⁷	0.21	0.18 (±0.022)	>10 ⁴
PFDPPS-Si	Drop cast	N/A	0.69	0.59 (±0.076)	>10 ⁶	0.12	0.11 (±0.006)	>10 ³
		220	1.05	0.85 (±0.120)	>10 ⁶	0.21	0.18 (±0.016)	>10 ⁵
	Solution sheared	N/A	0.82	0.63 (±0.112)	>10 ⁷	0.17	0.15 (±0.010)	>10 ³
		220	1.90	1.20 (±0.303)	>10 ⁶	0.30	0.23 (±0.032)	>10 ⁴

^a The OFETs were manufactured by using polymer films fabricated by drop casting and solution shearing methods. The *p*-channel and *n*-channel characteristics of ambipolar OFETs were measured with $V_{DS} = -100$ and $+100$ V, respectively. ^b The average mobilities were obtained from at least 15 OFET devices. ^c The thermal annealing was not applied. ^d The standard deviations of mobility values are presented in the parentheses.

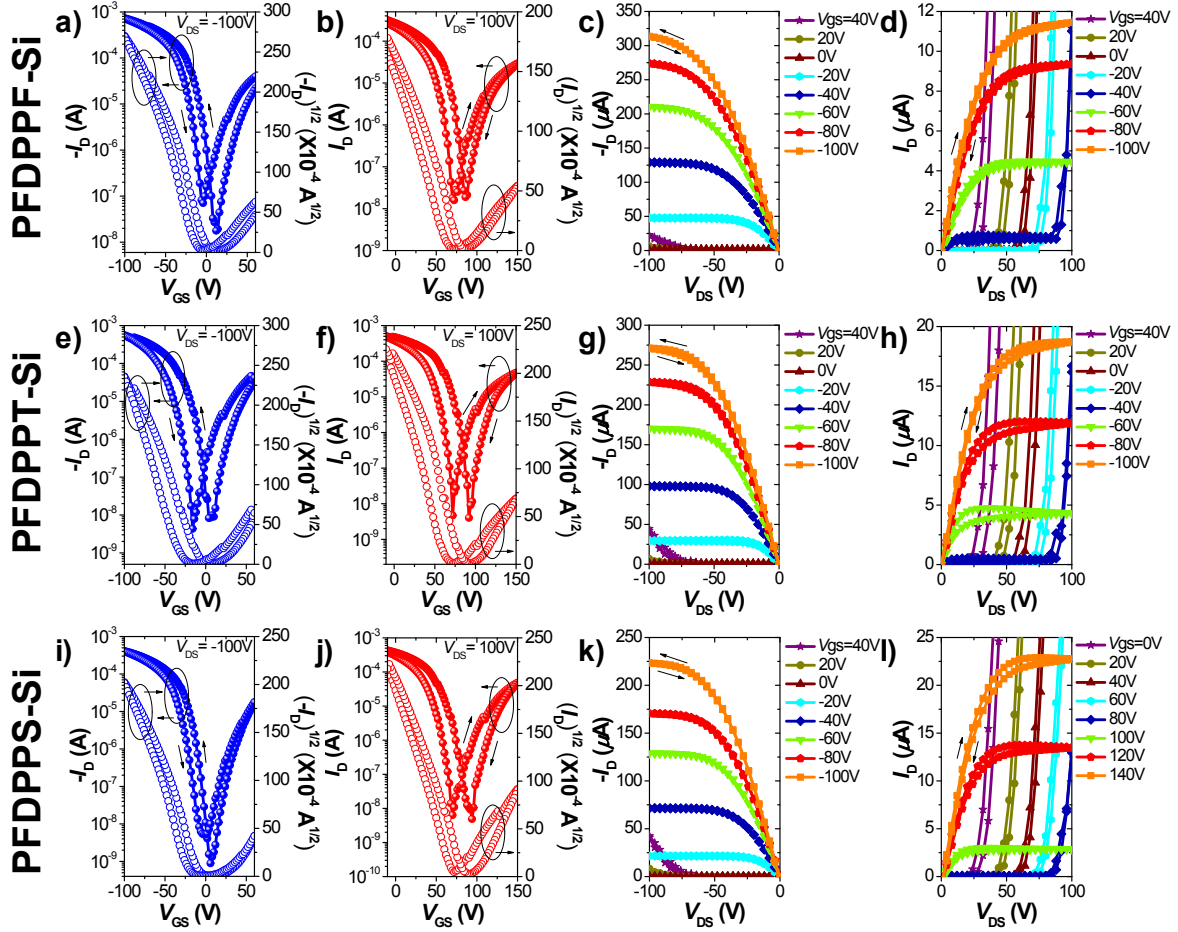


Fig. S5. I - V curves obtained from FET devices fabricated by optimized condition (solution shearing, thermal annealing at 220 °C): a-d) **PFDPFF-Si**, e-h) **PFDPPT-Si**, and i-l) **PFDPSS-Si**. Transfer characteristics at a, e, and i) for hole- and b, f, and j) for electron-enhancement operations with $V_{DS} = -100$ and $+100$ V, respectively. Output characteristics at c, g, and k) for p - and d, h, and l) for n -channel operation ($L = 50 \mu\text{m}$ and $W = 1000 \mu\text{m}$).

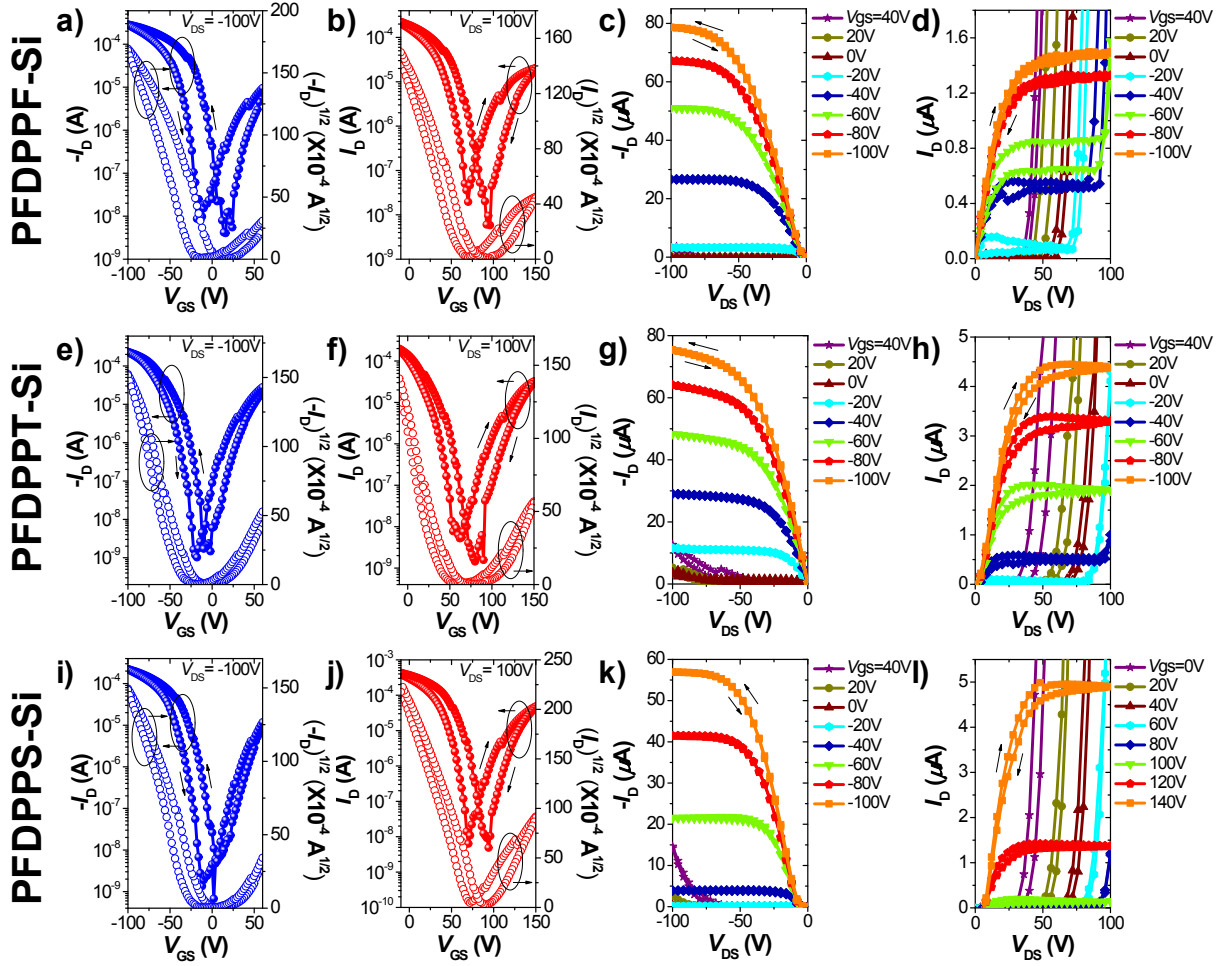


Fig. S6. I - V curves obtained from FET devices fabricated by solution shearing. Thermal annealing was not applied: a-d) **PFDPF-Si**, e-h) **PFDPT-Si**, and i-l) **PFDPPS-Si**. Transfer characteristics at a, e, and i) for hole- and b, f, and j) for electron-enhancement operations with $V_{DS} = -100$ and $+100$ V, respectively. Output characteristics at c, g, and k) for p - and d, h, and l) for n -channel operation ($L = 50 \mu\text{m}$ and $W = 1000 \mu\text{m}$).

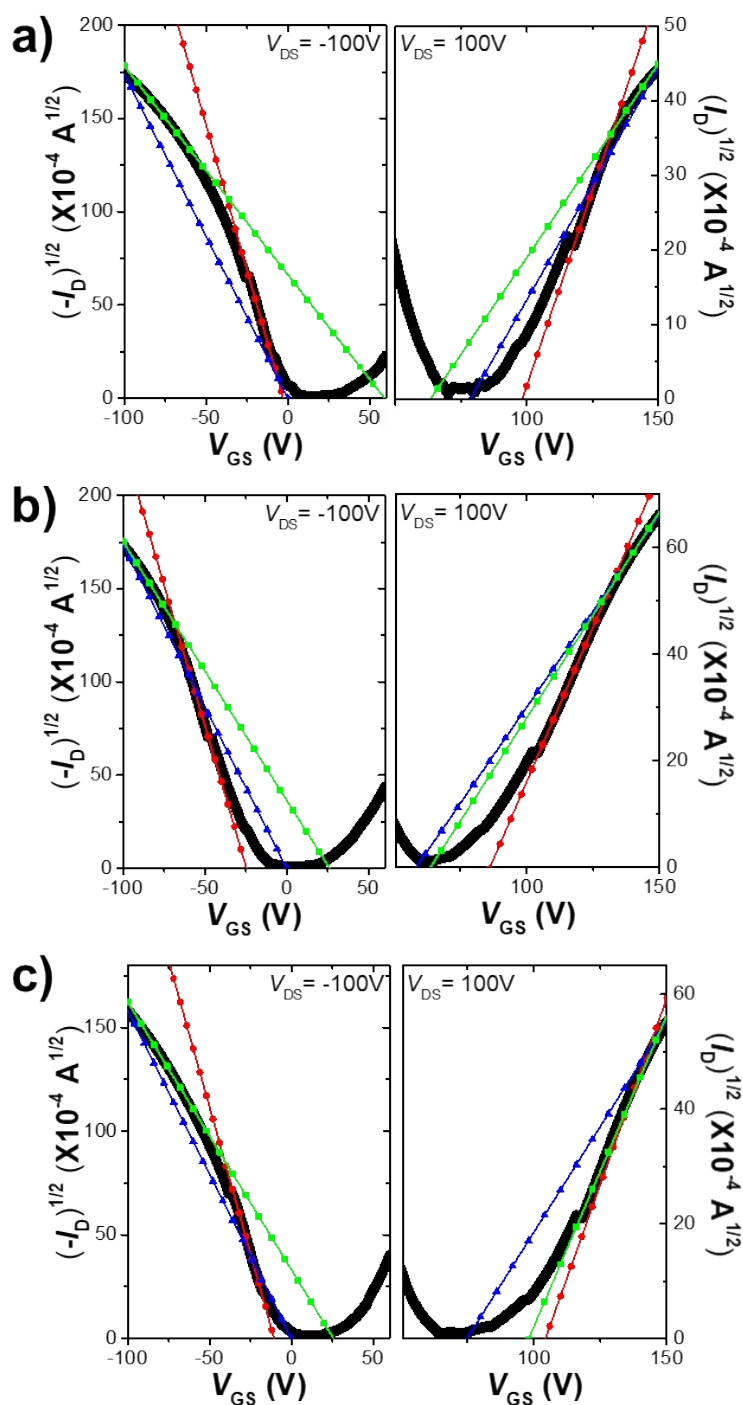


Fig. S7. *p*- and *n*-Channel transfer curves (left and right, respectively) of OFETs prepared by solution shearing with shearing speed of 0.1 mm s^{-1} . Thermal annealing was not applied: a) **PFDDPF-Si**, b) **PFDPPT-Si**, and c) **PFDDPPS-Si**. The red line is represented for the fitting line of the maximum mobility, the green line for the fitting line at a high gate voltage above the kink, and the blue line for the ideal FET characteristics which satisfy the ideal Shockley equations.

Table S5. Summary of the hole and electron mobilities and reliability factors of solution sheared, as-cast **PFDPPX-Si**-based OFETs.^a

Conditions ^a	<i>p</i> -Channel				<i>n</i> -Channel			
	μ_{max}^b [cm ² V ⁻¹ s ⁻¹]	μ_{highV}^c [cm ² V ⁻¹ s ⁻¹]	μ_{eff}^d [cm ² V ⁻¹ s ⁻¹]	R_{sat}^e	μ_{max} [cm ² V ⁻¹ s ⁻¹]	μ_{highV} [cm ² V ⁻¹ s ⁻¹]	μ_{eff} [cm ² V ⁻¹ s ⁻¹]	R_{sat}
PFDPPF-Si	1.20	0.17	0.32	0.27	0.12	0.05	0.04	0.33
PFDPPT-Si	0.94	0.30	0.30	0.31	0.14	0.08	0.05	0.38
PFDPPS-Si	0.82	0.21	0.24	0.29	0.15	0.12	0.05	0.34

^a The FET performance of the solution sheared **PFDPPX-Si**-based devices before annealing. ^b The maximum mobility of the FET devices. ^c The mobility obtained from the high gate voltage region above the kink of transfer curves. ^d The effective mobility from the equation, $\mu_{eff} = \mu_{max} \times R_{sat}$. ^e The reliability factor.