# **Electronic Supplementary Information**

# Tunable Light Harvesting Properties of a Highly Crystalline Alternating Terpolymer for High–Performing Solar Cells

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**Gel Permeation Chromatograms** 



M <sub>n</sub> (Da)	$M_{\rm w}$ (Da)	$M_{\rm P}$ (Da)	$M_{\rm z}$ (Da)	Polydispersity	% Area
94798	626553	287422	2586645	6.609313	100.00

Fig. S1 GPC chromatogram of PDPPPy.



M <sub>n</sub> (Da)	$M_{\rm w}$ (Da)	$M_{\rm P}$ (Da)	M <sub>z</sub> (Da)	Polydispersity	% Area
197790	1095020	930292	2557106	5.536283	100.00

Fig. S2 GPC chromatogram of PDPPPyT.



$M_{\rm n}$ (Da)	$M_{\rm w}$ (Da)	$M_{\rm P}$ (Da)	M <sub>z</sub> (Da)	Polydispersity	% Area
46558	90056	77361	155962	1.934260	100.00

Fig. S3 GPC chromatogram of PDPPT.

## Thermal analysis



**Fig. S4** DSC thermograms of PDPPPy (black), PDPPPyT (red), and PDPPT (blue). Heating cycle (solid line) and cooling cycle (dashed line).

### **GI-XRD** Data



**Fig. S5** Out-of-plane and in-plane profiles of XRD patterns. Sample: thermally annealed films of (a) PDPPPy, (b) PDPPPyT and (c) PDPPT. Out-of-plane (left side) and in-plane (right side).

Crystallographic parameters		Annealing Temperature				
		Pristine	150 °C	200 °C	250 °C	
Out-of-plane (100)	$q_{z}$ (Å-1)	0.2808	0.2686	0.2754	0.2764	
	d-spacing (Å)	21.43	22.29	21.73	21.76	
	FWHM (Å <sup>-1</sup> )	0.0351	0.0467	0.0315	0.0309	
In-plane (100)	$q_{z}$ (Å <sup>-1</sup> )	0.2810	0.2926	0.2829	0.2887	
	d-spacing (Å)	22.36	20.14	22.21	21.76	
	FWHM (Å <sup>-1</sup> )	-	-	-	-	

**Table S1** Crystallographic parameters calculated from the GI-XRD profiles of PDPPPy.

Crystallographic parameters		Annealing Temperature			
		Pristine	150 °C	200 °C	250 °C
	$q_{z}$ (Å <sup>-1</sup> )	-	-	-	0.2585
Out-of-plane (100)	d-spacing (Å)	-	-	-	23.41
	FWHM (Å-1)	-	-	-	0.0740
	$q_z$ (Å <sup>-1</sup> )	0.2596	0.2635	0.2635	0.2557
In-plane (100)	d-spacing (Å)	24.20	23.85	z23.85	24.57
	FWHM (Å-1)	0.1022	0.0712	0.0507	0.0338

**Table S2** Crystallographic parameters calculated from the GI-XRD profiles of PDPPPyT.

Crystallographic parameters		Annealing Temperature				
		Pristine	150 °C	200 °C	250 °C	
Out-of-plane (100)	$q_{z}$ (Å-1)	-	0.2347	0.2560	0.2414	
	d-spacing (Å)	-	25.49	23.46	25.49	
	FWHM (Å <sup>-1</sup> )	-	0.0619	0.0245	0.0407	
In-plane (100)	$q_{z}$ (Å <sup>-1</sup> )	0.2635	0.2616	0.2577	0.2616	
	d-spacing (Å)	23.39	24.02	24.39	24.02	
	FWHM (Å <sup>-1</sup> )	0.0437	0.0245	-	-	

**Table S3** Crystallographic parameters calculated from the GI-XRD profiles of PDPPT.

### **Output curves of the TFTs**



**Fig. S6** Output curves of (a) PDPPPy, (b) PDPPPyT, and (c) PDPPT TFTs. The range of the output curves is -40 to 20 V and  $V_{DS} = -100$  V.



Fig. S7. AFM images (size: 10  $\mu$ m X 10  $\mu$ m ) of photoactive layers. PDPPPy (a, d), PDPPPyT (b, e), and PDPPT (c, f). (a, b, c): the images obtained without DIO and (d, e, f): the images obtained with 3 vol% DIO.

The surface morphology of the active PSC layer was measured by AFM for better understanding the basis for the improved performance observed by adding DIO additives. As shown in Fig. S7, the surface morphology was observed to be different by adding 3 vol% DIO into the photoactive layers.