

## Supporting Information

### Highly Efficient Polymer Solar Cells *via* a Multiple Cascade Energy Level Engineering Strategy

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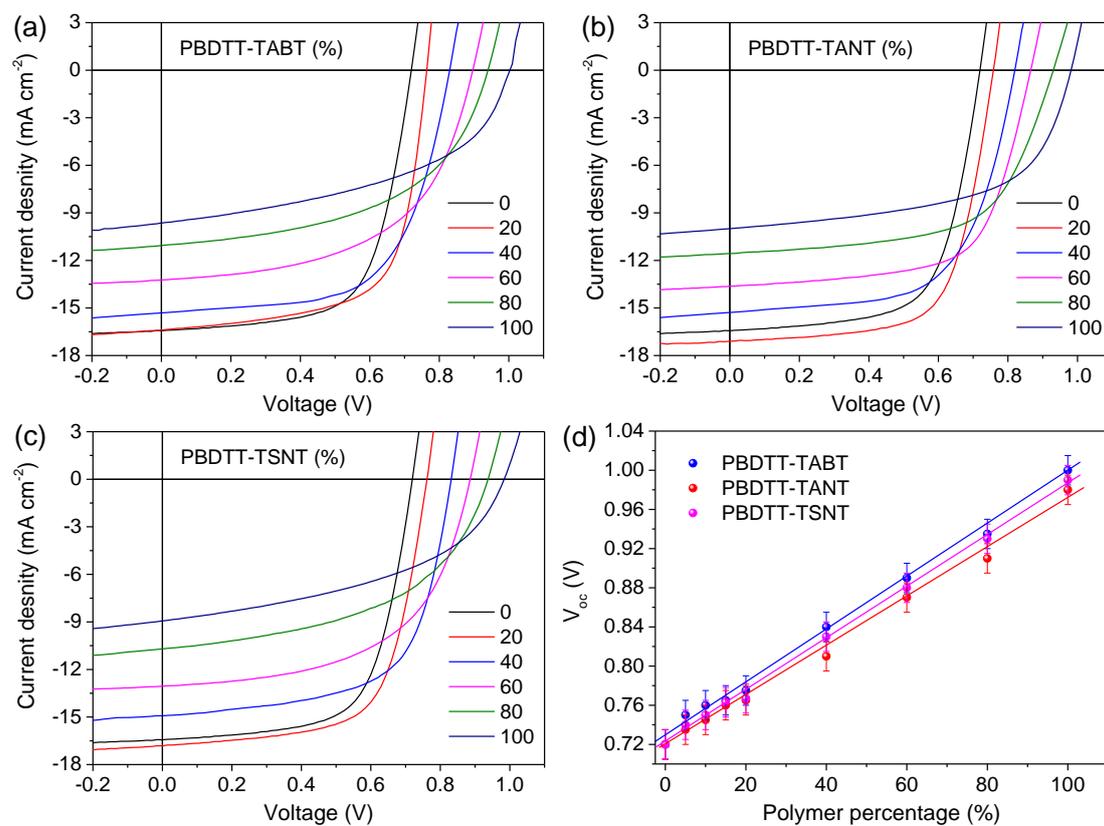
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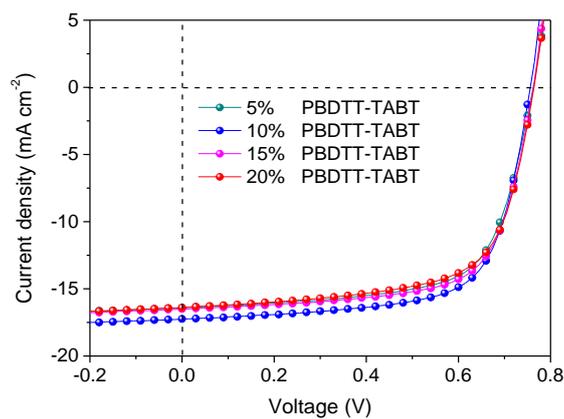
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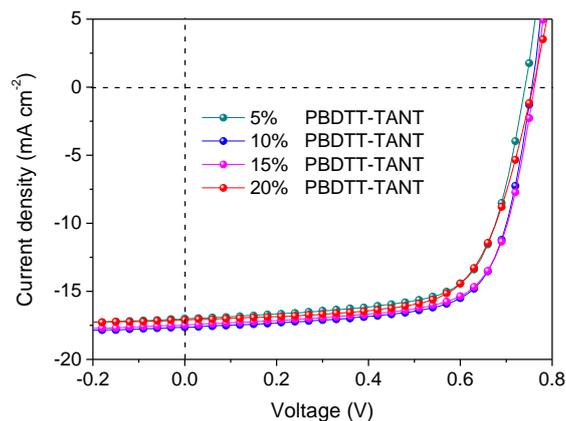
## 1. J-V curves PTB7:PC<sub>71</sub>BM with or without polymeric additives



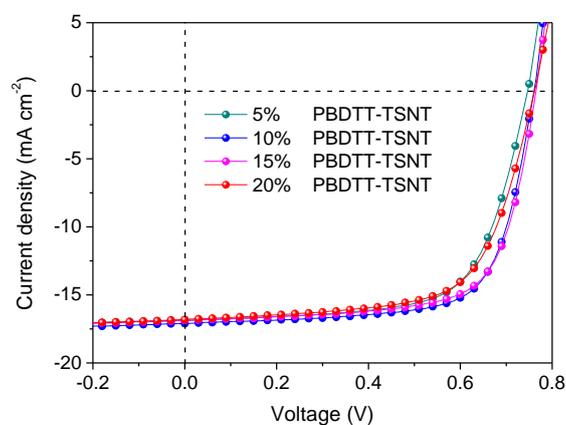
**Fig. S1** J-V curves of PTB7-based ternary blend PSCs with different amount of PBDTT-TABT (a), PBDTT-TANT (b), and PBDTT-TSNT (c). (d) Dependence of the V<sub>OC</sub> of the PSCs on the polymer additives content.



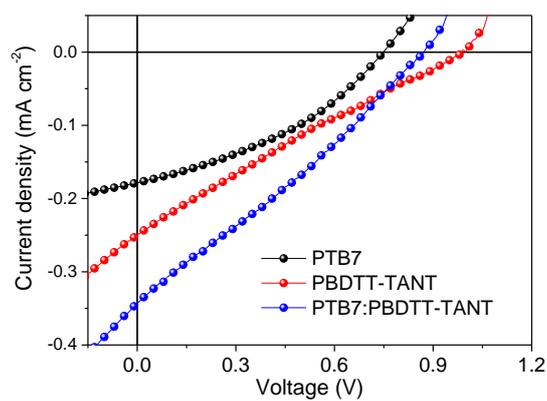
**Fig. S2** J-V curves of PBDTT-TABT-based ternary blend PSCs.



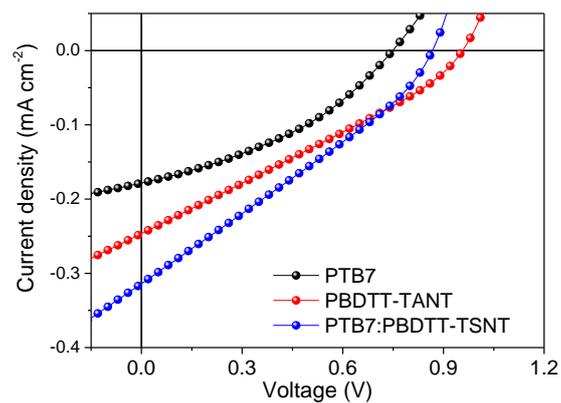
**Fig. S3** J-V curves of PBDTT-TANT-based ternary PSCs.



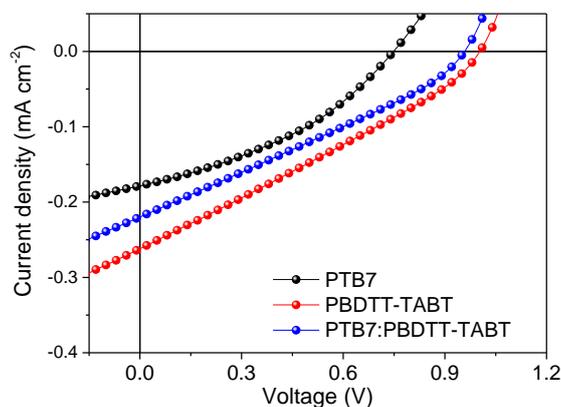
**Fig. S4** J-V curves of PBDTT-TSNT-based ternary blend PSCs.



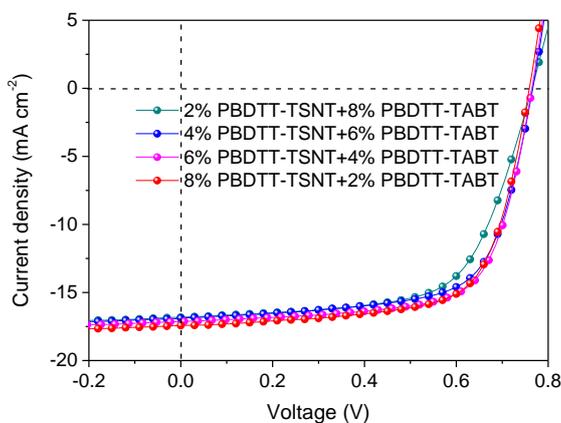
**Fig. S5** J-V curves of PSCs based on PTB7, PBDTT-TANT and PTB7:PBDTT-TANT (1:1) as active layer (without PC<sub>71</sub>BM).



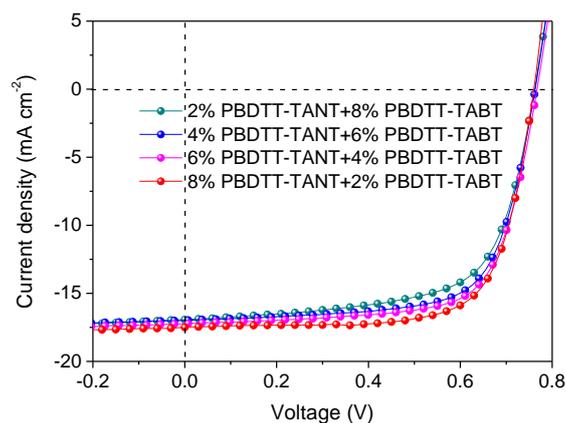
**Fig. S6** J-V curves of PSCs based on PTB7, PBDTT-TSNT and PTB7:PBDTT-TSNT (1:1) as active layer (without PC<sub>71</sub>BM).



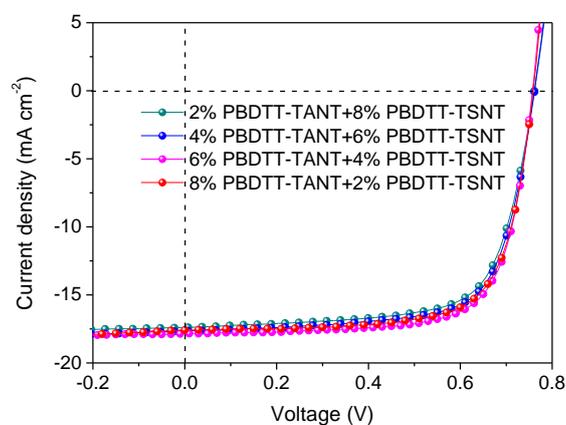
**Fig. S7** J-V curves of PSCs based on PTB7, PBDTT-TABT and PTB7:PBDTT-TABT (1:1) as active layer (without PC<sub>71</sub>BM).



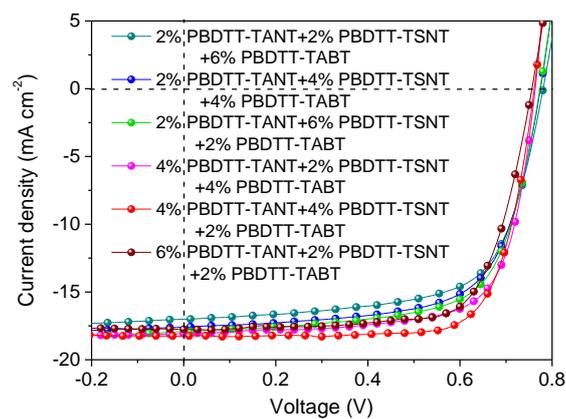
**Fig. S8** J-V curves of PBDTT-TSNT:PBDTT-TABT-based quaternary PSCs.



**Fig. S9** J-V curves of, PBDTT-TANT:PBDTT-TABT-based quaternary blend PSCs.



**Fig. S10** J-V curves of PBDTT-TANT:PBDTT-TSNT- based quaternary blend PSCs.



**Fig. S11** J-V curves of PTB7-based quintuple blend PSCs.

## 2. Performance summary of PTB7:PC<sub>71</sub>BM with or without additional copolymers.

**Table S1** Device data of the multiple blend PSCs.

Blend type	Donor composition	V <sub>oc</sub> (V)	J <sub>sc</sub> (mA·cm <sup>-2</sup> )	FF (%)	PCE <sub>max/ave</sub> (%)	R <sub>s</sub> (Ω·cm <sup>2</sup> )	R <sub>sh</sub> (Ω·cm <sup>2</sup> )
Binary blend <sup>[a]</sup>	100% PTB7	0.720	15.87	65.6	7.50/7.24	8.7	806.9
	100% PBDTT-TABT	0.975	11.12	62.1	6.74/6.61	12.8	494.2
	100% PBDTT-TANT	0.945	13.06	65.2	8.04/7.84	10.8	629.2
	100% PBDTT-TSNT	0.975	10.32	60.9	6.13/5.96	14.7	601.8
Ternary blend	5% PBDTT-TABT	0.765	16.41	67.4	8.46/8.20	6.3	548.0
	5% PBDTT-TANT	0.735	17.00	69.2	8.65/8.54	5.2	467.8
	5% PBDTT-TSNT	0.750	16.94	67.6	8.59/8.39	6.1	1176.2
	10% PBDTT-TABT	0.765	16.91	69.3	8.96/8.64	4.3	933.5
	10% PBDTT-TANT	0.750	17.64	70.7	9.35/9.00	4.6	1308.4

	10% PBDTT-TSNT	0.765	17.11	70.2	9.18/8.77	4.2	1203.8
	15% PBDTT-TABT	0.765	16.50	68.4	8.63/8.34	4.4	794.3
	15% PBDTT-TANT	0.765	17.48	69.3	9.27/8.97	4.2	473.3
	15% PBDTT-TSNT	0.765	16.87	69.9	9.03/8.68	4.3	1077.4
	20% PBDTT-TABT	0.765	16.4	66.4	8.33/8.12	4.7	443.3
	20% PBDTT-TANT	0.765	17.10	66.5	8.70/8.55	6.4	683.6
	20% PBDTT-TSNT	0.765	16.82	65.6	8.44/8.29	6.4	621.1
Quaternary blend	2% PBDTT-TSNT	0.765	16.82	65.9	8.32/8.06	9.0	786.6
	+8% PBDTT-TABT						
	4% PBDTT-TSNT	0.765	16.89	68.2	8.81/8.530	6.2	634.9
	+6% PBDTT-TABT						
	6% PBDTT-TSNT	0.765	17.13	69.9	9.16/8.88	6.0	857.7
	+4% PBDTT-TABT						

8% PBDTT-TSNT	0.765	17.43	69.5	9.27/9.01	6.3	1587.0
+2% PBDTT-TABT						
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2% PBDTT-TANT	0.765	16.93	67.2	8.53/8.26	6.8	623.5
+8% PBDTT-TABT						
4% PBDTT-TANT	0.765	16.98	69.8	9.07/8.39	6.0	791.6
+6% PBDTT-TABT						
6% PBDTT-TANT	0.761	17.22	70.8	9.28/9.00	5.9	815.5
+4% PBDTT-TABT						
8% PBDTT-TANT	0.750	17.50	72.9	9.58/9.24	5.8	1842.4
+2% PBDTT-TABT						
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2% PBDTT-TANT	0.765	17.38	70.1	9.28/9.07	5.5	824.8
+8% PBDTT-TSNT						
4% PBDTT-TANT	0.750	17.64	70.8	9.36/9.11	5.2	859.9

	+6% PBDTT-TSNT						
	6% PBDTT-TANT	0.750	17.83	73.5	9.83/9.55	5.1	1900.1
	+4% PBDTT-TSNT						
	8% PBDTT-TANT	0.750	17.58	72.9	9.61/9.36	5.3	1334.0
	+2% PBDTT-TSNT						
	2% PBDTT-TANT+2%						
	PBDTT-TSNT+6% PBDTT-TABT	0.780	17.01	66.6	8.84/8.61	6.5	466.0
	2% PBDTT-TANT+4%						
Quintuple	PBDTT-TSNT+4% PBDTT-TABT	0.765	17.59	67.9	9.14/8.87	6.5	625.5
blend	2% PBDTT-TANT+6%						
	PBDTT-TSNT+2% PBDTT-TABT	0.765	17.93	68.5	9.39/9.04	6.2	525.9
	4% PBDTT-TANT+2%						
	PBDTT-TSNT +4% PBDTT-TABT	0.761	18.06	71.9	9.88/9.58	4.9	612.2

4% PBDTT-TANT+4%	0.756	18.24	75.4	10.40/9.95	5.1	2468.6
PBDTT-TSNT+2% PBDTT-TABT						
6% PBDTT-TANT+2%	0.750	17.78	72.1	9.60/9.51	6.0	1527.0
PBDTT-TSNT+2% PBDTT-TABT						

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[a] The photovoltaic parameters of PBDTT-TABT-, PBDTT-TANT- and PBDTT-TSNT-based binary blend PSCs came from our previous work [ref. 1].

## References

1 X.P. Xu, K. Feng, K. Li, Q. Peng, *J. Mater. Chem. A*, 2015, **3**, 23149-23161.