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Electronic Supplementary Information For:

Efficient and stable organic solar cells via sequential process

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Table s1 Average and best device data based on s-BHJ films with different thicknesses and different DCB content in mixed solvent

Donor	Acceptor	Active layer	DCB	V_{OC}	J_{SC}	Calc J_{SC}	FF	PCE (%)	
(nm)	(nm)	(nm)	(%)	(V)	(mA cm^{-2})	(mA cm^{-2})	(%)	ave	best
50	35	76	5	0.81	14.2	13.9	61.9	7.1	7.3
65	35	90	5	0.81	15.3	14.9	60.4	7.5	7.8
80	35	103	5	0.80	15.3	15.0	55.8	6.9	7.1
65	50	105	5	0.82	14.9	14.6	64.1	7.8	8.0
80	/[a]	/[a]	0	0.75	3.91	3.75	35.6	1.1	1.1
80	50	123	3	0.82	16.6	16.3	61.8	8.4	8.6
80	50	120	5	0.81	16.6	16.4	62.7	8.5	8.6
80	50	117	7	0.81	16.7	16.5	62.4	8.4	8.6
95	50	134	5	0.81	16.1	15.4	60.5	7.8	8.1

[a] $PC_{71}BM$ can't form a uniform film with pure DCM.

Table s2 Compositions of m-BHJ and s-BHJ films in different depth

Structure	DIO (%)	Depth (nm)	Atom ratio of O/F	Polymer weight content (%)
m-BHJ	0	0	3.037	62
m-BHJ	0	24	4.769	38
m-BHJ	0	48	6.022	30
m-BHJ	0	72	4.506	40
m-BHJ	0	96	3.393	55
m-BHJ	0	120	4.135	44
m-BHJ	3	0	4.001	46
m-BHJ	3	24	5.488	33
m-BHJ	3	48	4.008	46
m-BHJ	3	72	3.743	49
m-BHJ	3	96	2.907	65
m-BHJ	3	120	3.355	56
s-BHJ	0	0	20.61	8.4
s-BHJ	0	24	4.171	44
s-BHJ	0	48	3.323	56
s-BHJ	0	72	2.770	69
s-BHJ	0	96	2.513	74
s-BHJ	0	120	2.378	82

Table s3 Hole and electron mobilities of m-BHJ and s-BHJ films

Device	DIO (%)	Thickness (nm)	Pre-annealing	Stability test	μ_h ($cm^2V^{-1}s^{-1}$)	μ_e ($cm^2V^{-1}s^{-1}$)
m-BHJ-0	0	97	No	No	1.9×10^{-4}	2.6×10^{-4}
m-BHJ-1	3	100	No	No	3.2×10^{-4}	2.8×10^{-4}
m-BHJ-2	3	101	No	Yes	4.1×10^{-5}	3.2×10^{-4}
m-BHJ-3	3	102	Yes	No	1.2×10^{-4}	3.0×10^{-4}
m-BHJ-4	3	102	Yes	Yes	5.3×10^{-5}	3.2×10^{-4}
s-BHJ-1	0	120	No	No	8.1×10^{-4}	2.8×10^{-4}
s-BHJ-2	0	122	No	Yes	1.4×10^{-4}	2.9×10^{-4}
s-BHJ-3	0	118	Yes	No	7.9×10^{-4}	2.7×10^{-4}
s-BHJ-4	0	121	Yes	Yes	8.1×10^{-4}	2.9×10^{-4}

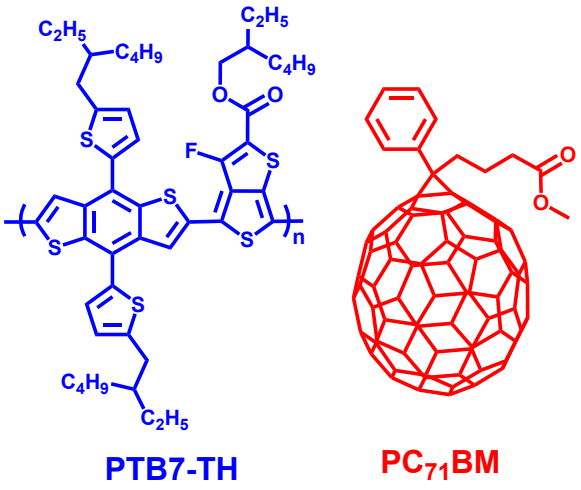

Fig. s1 Chemical structures of PTB7-TH and $PC_{71}BM$.

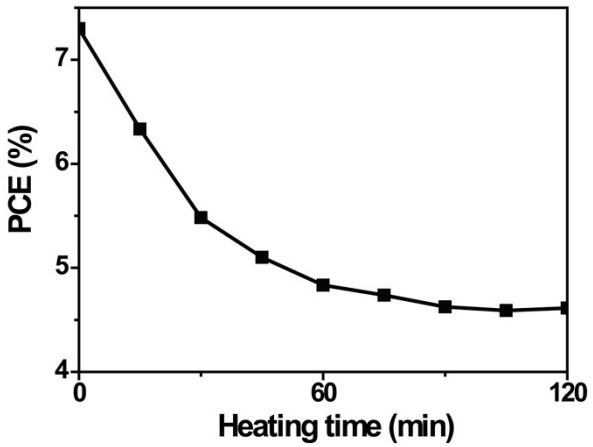

Fig. s2 Stability curve of the m-BHJ-0 device under continuous heating at 130 °C for 120 min.

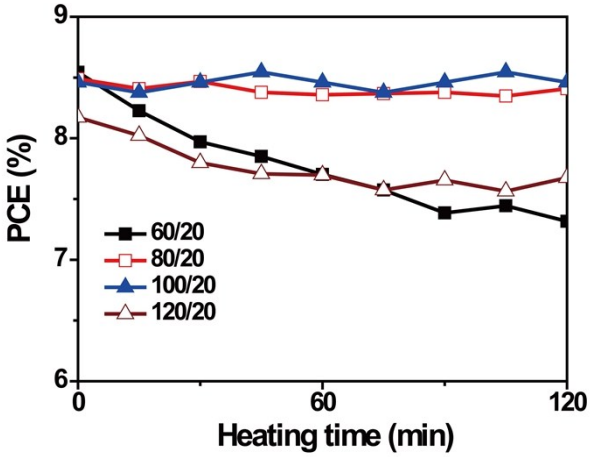

Fig. s3 Stability curves of s-BHJ devices with different pre-annealing temperatures.

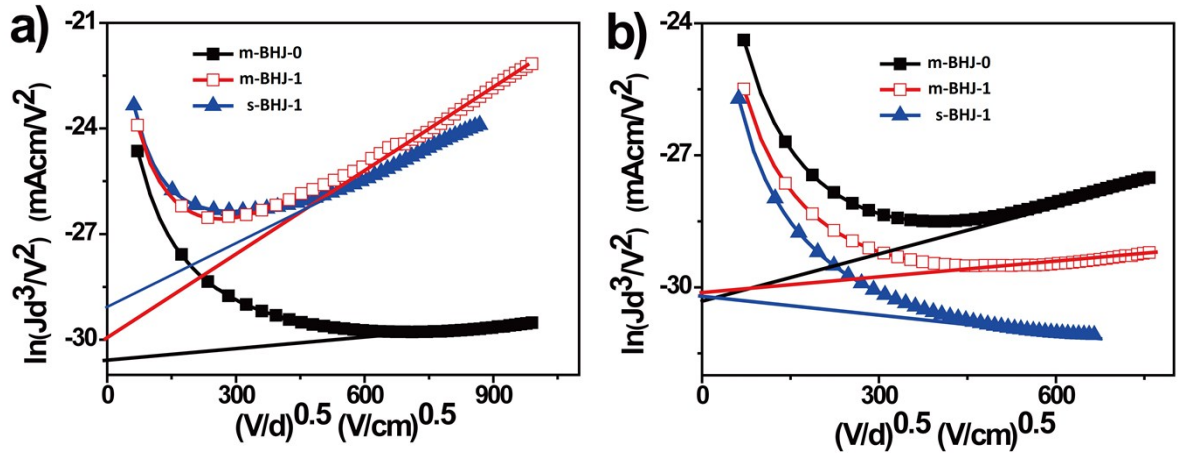

Fig. s4 *J-V* characteristics under dark for a) hole-only and b) electron-only devices based on m-BHJ and s-BHJ films.

Calculation of the surface compositions

$$\text{PTB7-TH weight content} = \frac{\textit{Polymer weight}}{\textit{Polymer weight} + \textit{PCBM weight}}$$

$$= \frac{n_{polymer} M_{polymer}}{n_{polymer} M_{polymer} + n_{PCBM} M_{PCBM}}$$

$$= \frac{1}{1 + \frac{n_{PCBM} M_{PCBM}}{n_{polymer} M_{polymer}}}$$

$$\frac{2n_{polymer} + 2n_{PCBM}}{n_{polymer}} = \frac{O}{F}$$

$$\frac{n_{PCBM}}{n_{polymer}} = 0.5\left(\frac{O}{F} - 2\right) = 0.5\frac{O}{F} - 1$$

$$\text{PTB7-TH weight content} = \frac{1}{\left(\frac{M_{PCBM}}{M_{polymer}}\right)\left(0.5\frac{O}{F} - 1\right) + 1}$$

M_{PCBM} is the molecular weight of $PC_{71}BM$ (1016);

$M_{Polymer}$ is the molecular weight of repeat unit of PTB7-TH (905);

n_{PCBM} is the mole number of $PC_{71}BM$;

$n_{Polymer}$ is the mole number of repeat unit of PTB7-TH;

$\frac{O}{F}$ is the atom ratio of O/F.