

## Supplementary Information

### Restricting the Liquid-Liquid Phase Separation of PTB7-Th:PF12TBT:PC<sub>71</sub>BM by Enhanced PTB7-Th Solution Aggregation to Optimize the Interpenetrating Network

Bin Tang<sup>a,b</sup>, Jiangang Liu<sup>\*a</sup>, Xinxiu Cao<sup>a</sup>, Qiaoqiao Zhao<sup>a</sup>, Xinhong Yu<sup>a</sup>, Shijun  
Zheng<sup>b</sup> and Yanchun Han<sup>\*a</sup>

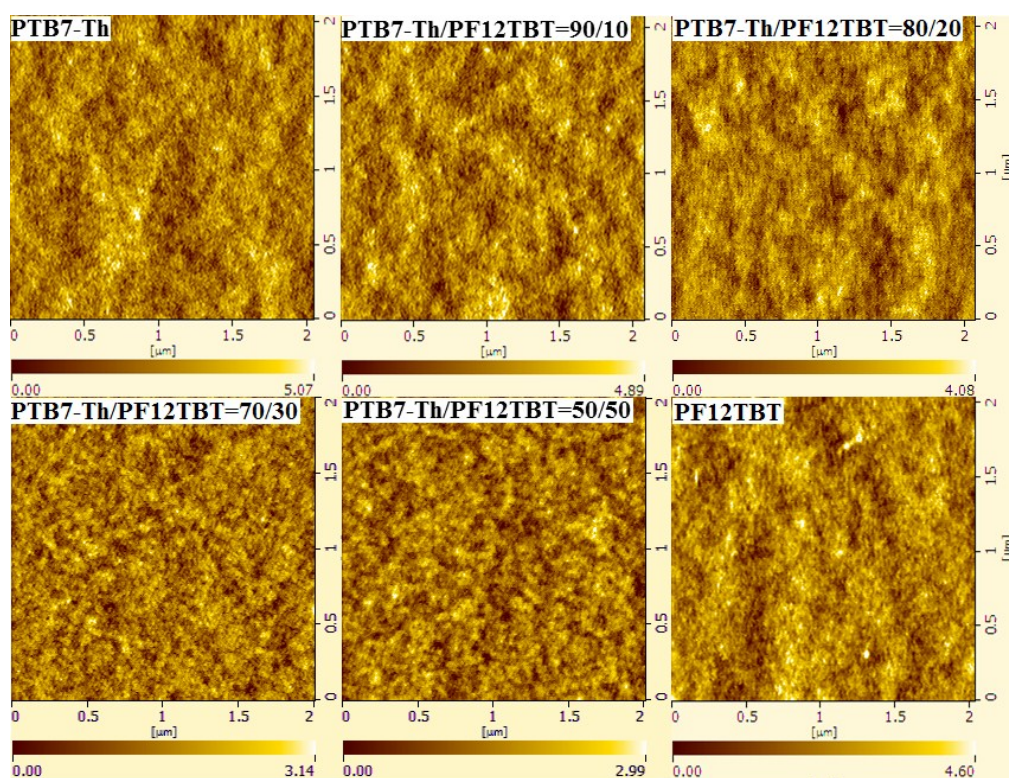
<sup>a</sup>State Key Laboratory of Polymer Physics and Chemistry, Changchun Institute of  
Applied Chemistry, Chinese Academy of Sciences, 5625 Renmin Street, Changchun  
130022, P. R. China;

<sup>b</sup>School of Materials Science and Engineering, Zhengzhou University, Zhengzhou  
450001, P. R. China

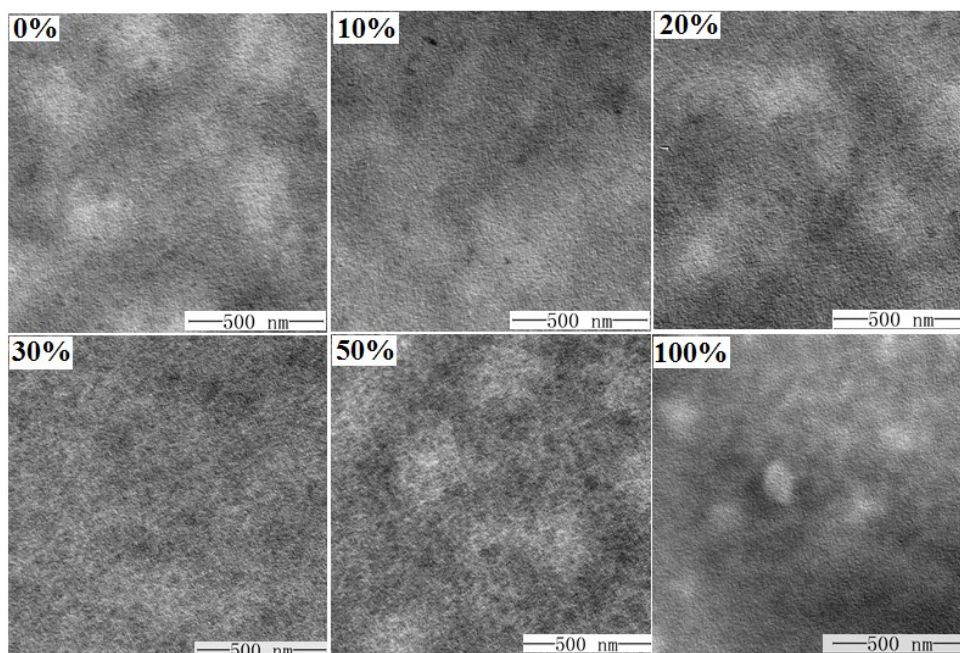
\* To whom correspondence should be addressed

Email: [ychan@ciac.ac.cn](mailto:ychan@ciac.ac.cn) (Yanchun Han); [niitawh@ciac.ac.cn](mailto:niitawh@ciac.ac.cn) (Jiangang Liu)

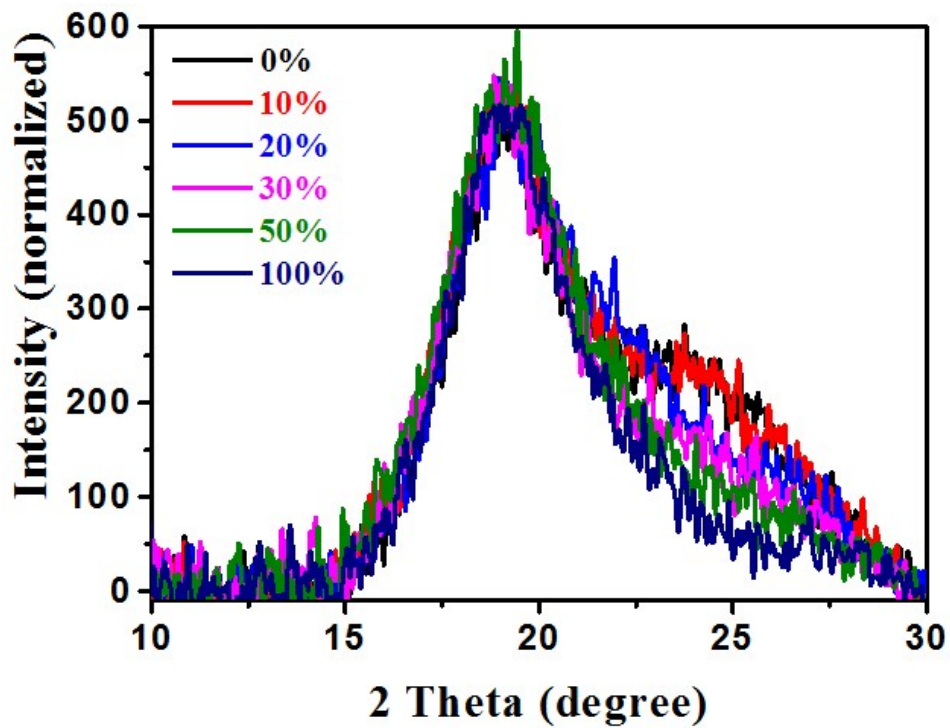
To check the compatibility between PTB7-Th and PF12TBT, the morphology of PTB7-Th:PF12TBT films with different blend ratios were checked by AFM and the AFM height images were shown in Fig. S1. The films showed smooth surface and no large phase separation was observed, suggesting the good compatibility between PTB7-Th and PF12TBT.



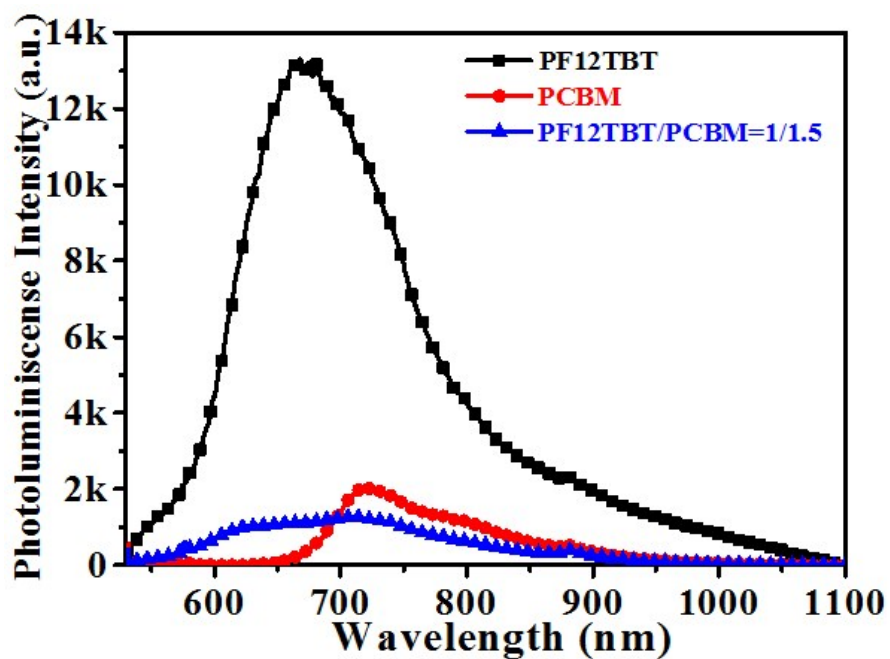
**Figure S1** The AFM height images of PTB7-Th:PF12TBT films with different blend ratios.



**Figure S2** The TEM images of PTB7-Th:PF12TBT:PC<sub>71</sub>BM films with increased PF12TBT loading ratio. Increased phase separation was observed with the rise of PF12TBT loading ratio. The PTB7-Th:PC<sub>71</sub>BM sample showed a featureless morphology which is in agreement with previous report.



**Figure S3** The out of plane GIXD profile of PTB7-Th:PF12TBT:PC<sub>71</sub>BM films with increased PF12TBT loading ratio. The (010) peak intensity of PTB7-Th decreased with increased PF12TBT content.



**Figure S4** The Steady state PL spectra of PF12TBT, PC<sub>71</sub>BM and PF12TBT:PC<sub>71</sub>BM films when excited at 532 nm. The highly quenched PF12TBT emission in PF12TBT:PC<sub>71</sub>BM films may further suggest that PF12TBT tend to be embedded in PC<sub>71</sub>BM domains.

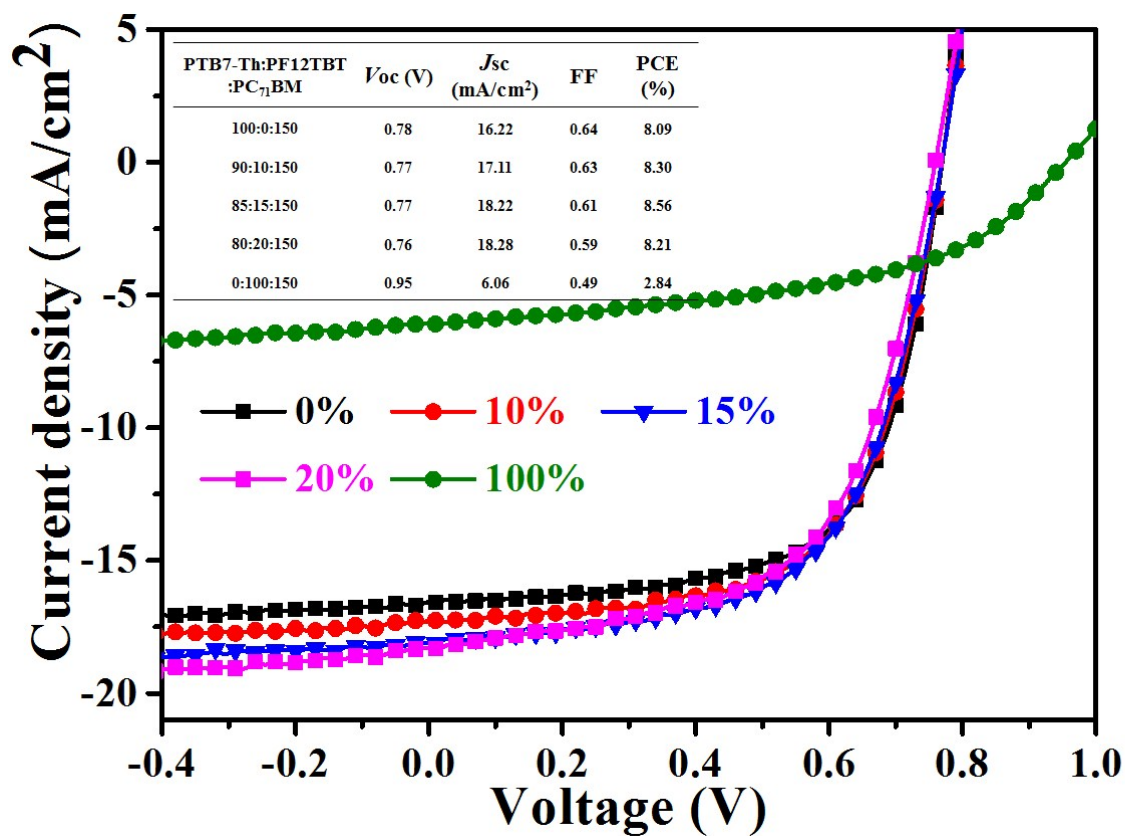
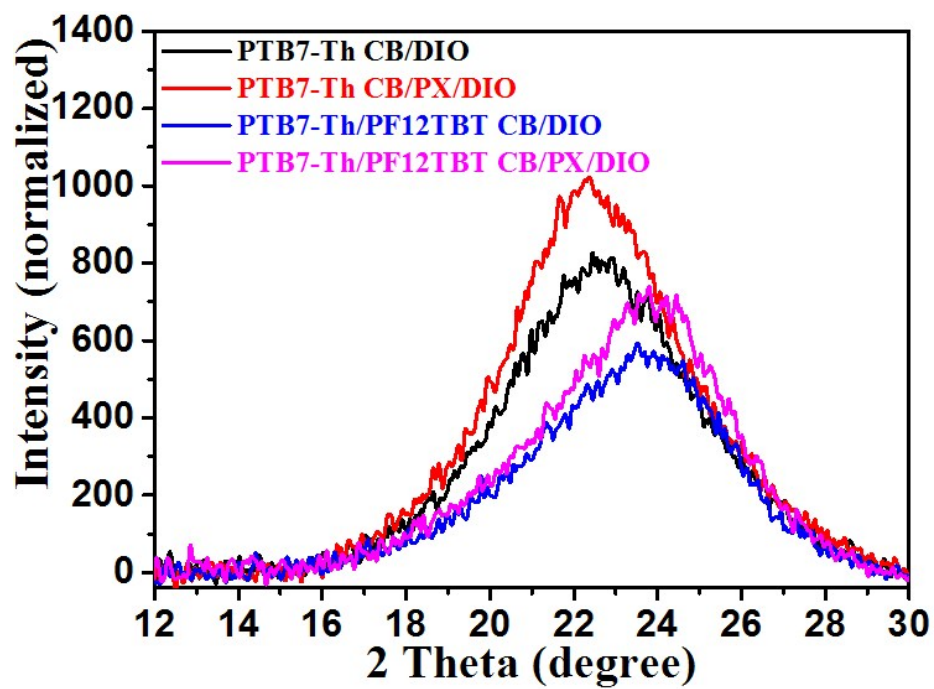
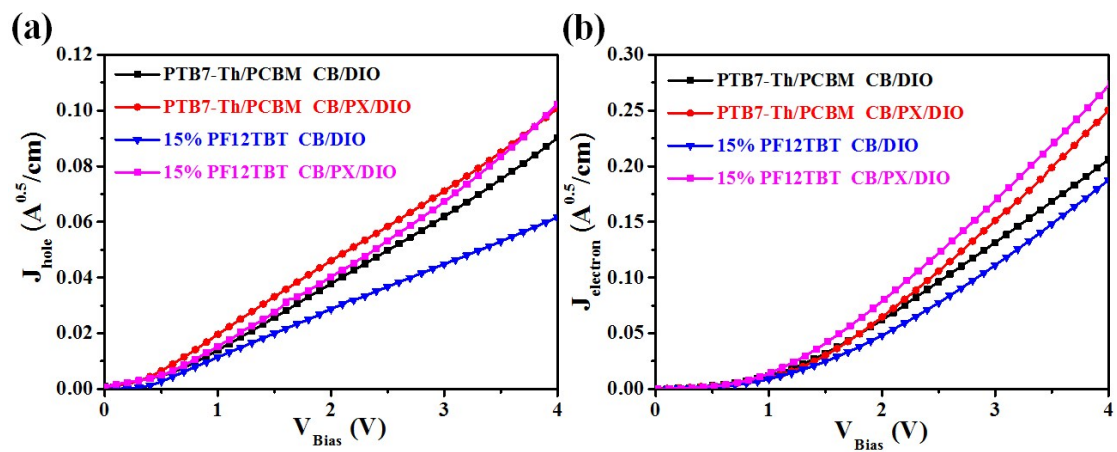


Figure S5 The current density *versus* voltage curves of PTB7-Th:PF12TBT:PC<sub>71</sub>BM devices with different contents of PF12TBT.



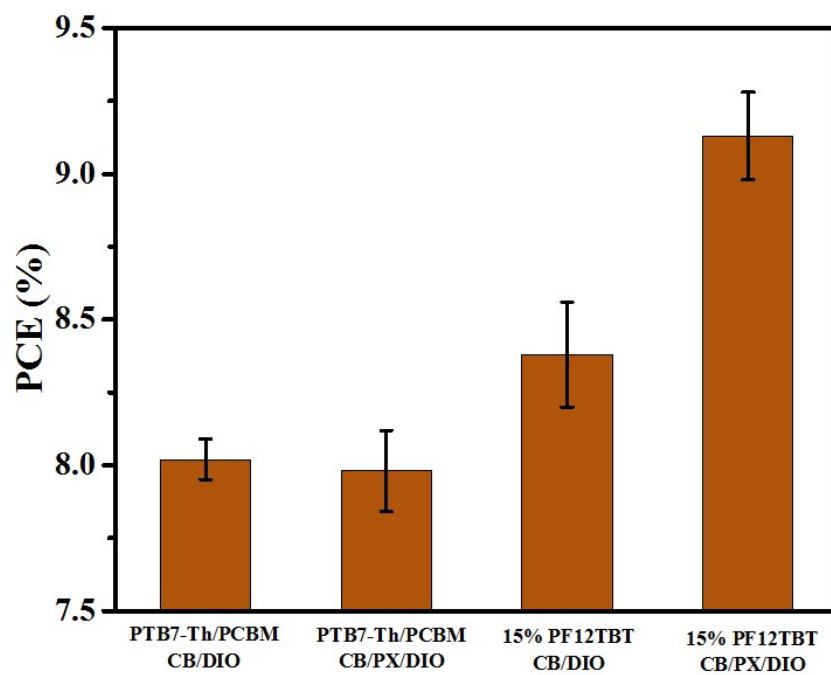
**Figure S6** The out of plane GIXD profile for the PTB7-Th and PTB7-Th:PF12TBT films processed from CB/DIO or CB/PX/DIO.



**Figure S7** Square root of hole current densities *versus* bias voltage of the hole only devices **(a)**.

Square root of electron current densities *versus* bias voltage of the electron only devices **(b)**.





**Figure S8** Power conversion efficiencies of binary and ternary devices processed from CB/DIO or CB/PX/DIO. The average value and error were obtained from ten devices.

**Table S1** The contact angle of different solvents on PTB7-Th, PC<sub>71</sub>BM and PF12TBT films and calculated surface energy.

	Contact angle (degree)		Surface energy (mN m <sup>-1</sup> )
	ultrapure water	ethylene glycol	
PTB7-Th	100.2	75.1	23.5
PC <sub>71</sub> BM	79.8	53.7	29.7
PF12TBT	98.3	66.3	33.1