Supporting Information

Mechanically-Robust and High-Performance Ternary Solar Cells Combining the Merits of All-Polymer and Fullerene Blends

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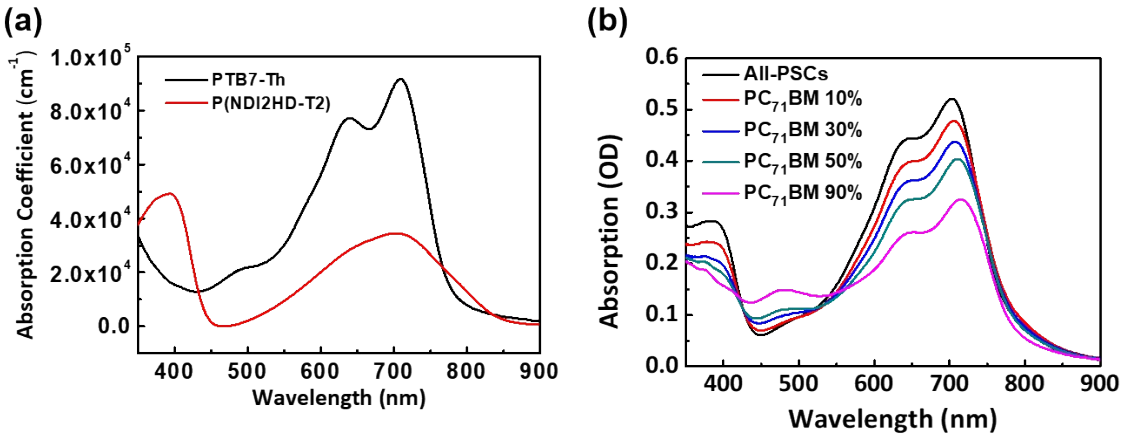

Figure S1. (a) UV-vis absorption spectra of pristine films of PTB7-Th and P(NDI2HD-T2). (b) UV-vis absorption spectra of all-PSC and ternary blends with different $PC_{71}BM$ contents.

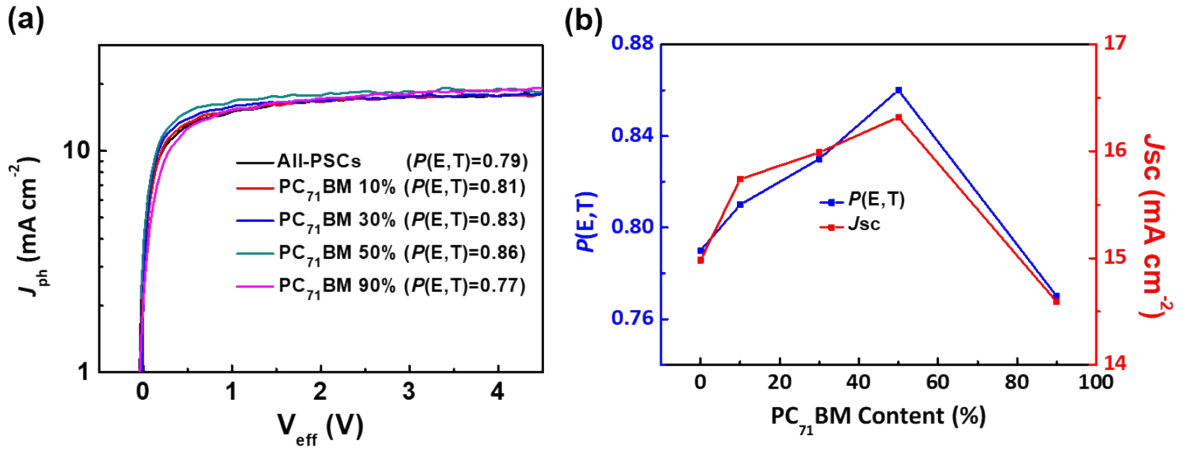

Figure S2. (a) Photocurrent analysis and (b) P(E,T) and J_{SC} values of all-PSCs and ternary-PSCs as a function of $PC_{71}BM$ content.

Table S1. SCLC mobility values of binary and ternary blend films.

PTB7-Th:P(NDI2HD-T2):$PC_{71}BM$	μ_h [$cm^2\ V^{-1}s^{-1}$]	μ_e [$cm^2\ V^{-1}s^{-1}$]
1.3:1:0 (All-PSCs)	1.2×10^{-4}	3.1×10^{-5}
1.3:0.9:0.1 ($PC_{71}BM$ 10%)	6.4×10^{-5}	1.8×10^{-5}
1.3:0.7:0.3 ($PC_{71}BM$ 30%)	8.5×10^{-5}	1.7×10^{-5}
1.3:0.5:0.5 ($PC_{71}BM$ 50%)	5.9×10^{-5}	1.8×10^{-5}
1.3:0.1:0.9 ($PC_{71}BM$ 90%)	4.9×10^{-5}	2.4×10^{-5}
1:0:1.5 ($PC_{71}BM$-PSCs)	2.2×10^{-4}	1.2×10^{-4}

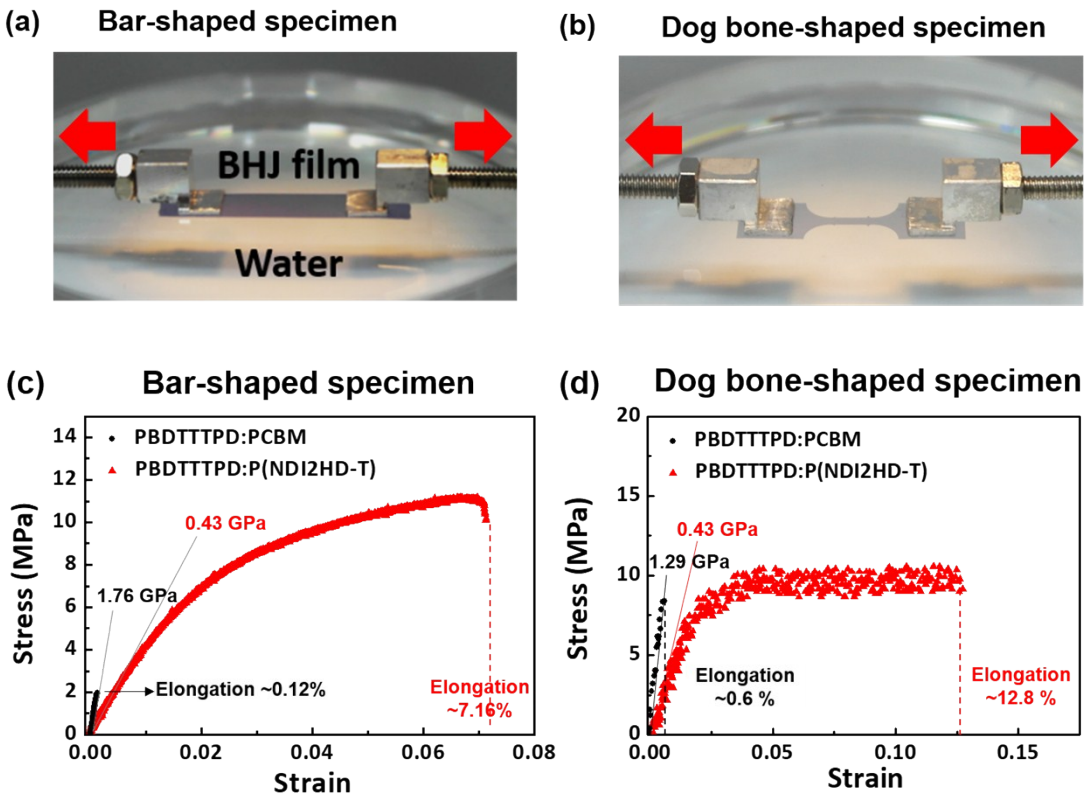

Figure S3. Photographs of (a) a bar-shaped specimen showing inhomogeneous strain distribution near the grips and (b) a dog bone-shaped specimen under tensile stress. Stress-strain curves of free-standing BHJ active layer blend films performed on (c) bar-shaped specimens and (d) dog bone-shaped specimens.

We examined the effects of specimen shape on the mechanical properties of BHJ films by using PBDTTTPD:PCBM and PBDTTTPD:P(NDI2HD)-T blend films for which the mechanical properties with bar-shaped specimens were investigated in our previous work with bar-shaped specimens.[1] We obtained the stress-strain curves of the dog bone-shaped free-standing films of the PBDTTTPD:PCBM and PBDTTTPD:P(NDI2HD-T) blends and compared them with those of the bar-shaped films (Figure S3). When the shape of specimen was changed from the bar type to the dog bone type, the values of elongation at break in both the PBDTTTPD:PCBM and PBDTTTPD:P(NDI2HD-T) blends increased from 0.12% (for the PCBM-PSC film) and 7.16% (for the all-PSC film) to 0.6% to 12.8%, respectively, while the values of tensile modulus remained constant, demonstrating the marked improvement in the tensile stress measuring system. Experiments performed with both the bar- and dog bone-shaped specimens clearly demonstrated the superior mechanical properties of the all-PSC films relative to those of the PCBM-PSC films. The M_n and $Đ$ of the PBDTTTPD and P(NDI2HD-T) polymers were measured to be 22 kg/mol and 2.02, and 48 kg/mol and 2.11, respectively. These sample conditions employed for the mechanical characterization in Figure S3 are almost identical to those used to prepare binary blend films used in our previous work.[1]

Table S2. Summary of elastic modulus, crack onset strain, and toughness values of all-PSC binary and ternary blend films.

Blend films	Elastic modulus (GPa)	Crack onset strain (%)	Toughness (J m^{-3})
All-PSCs	0.98 ± 0.04	11.6 ± 1.3	2237 ± 261
$PC_{71}BM$ 10%	1.13 ± 0.11	10.6 ± 0.7	1939 ± 158
$PC_{71}BM$ 30%	1.29 ± 0.09	10.7 ± 0.7	2160 ± 162
$PC_{71}BM$ 50%	1.50 ± 0.04	7.6 ± 0.4	1742 ± 112
$PC_{71}BM$-PSCs	2.64 ± 0.12	1.1 ± 0.2	154 ± 35

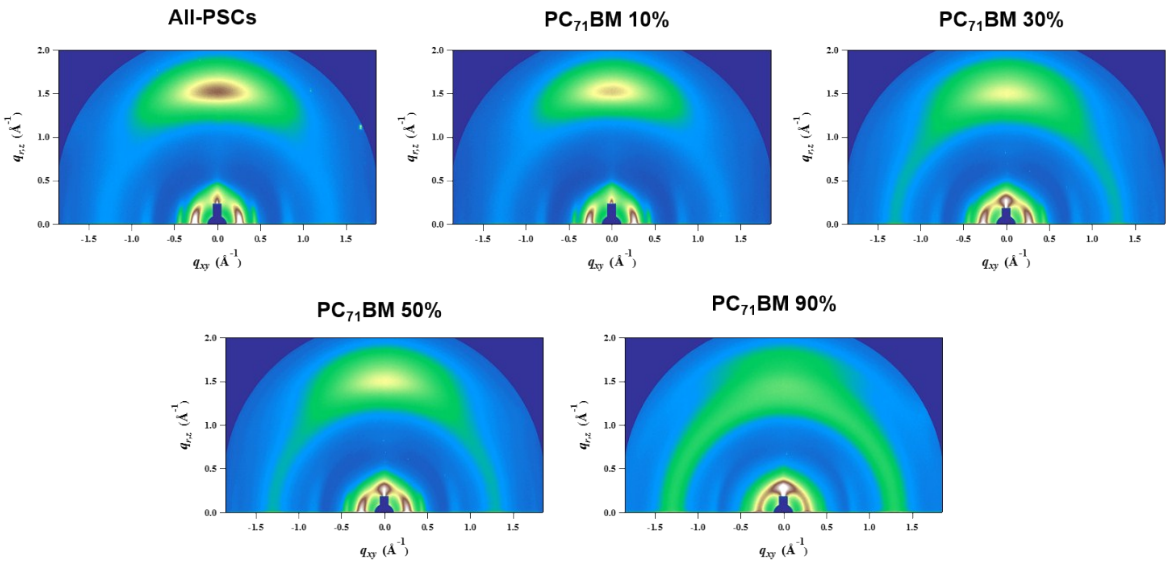

Figure S4. 2D-GIXS patterns of all-PSC binary and ternary blend films.

References

[1] T. Kim, J.-H. Kim, T. E. Kang, C. Lee, H. Kang, M. Shin, C. Wang, B. Ma, U. Jeong, T.-S. Kim, B. J. Kim, *Nat. Commun.* 2015, **6**, 8547