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Supplementary Information

Flexible and highly efficient perovskite solar cells with a large active area incorporating cobalt-doped poly(3-hexylthiophene) for enhanced open-circuit voltage

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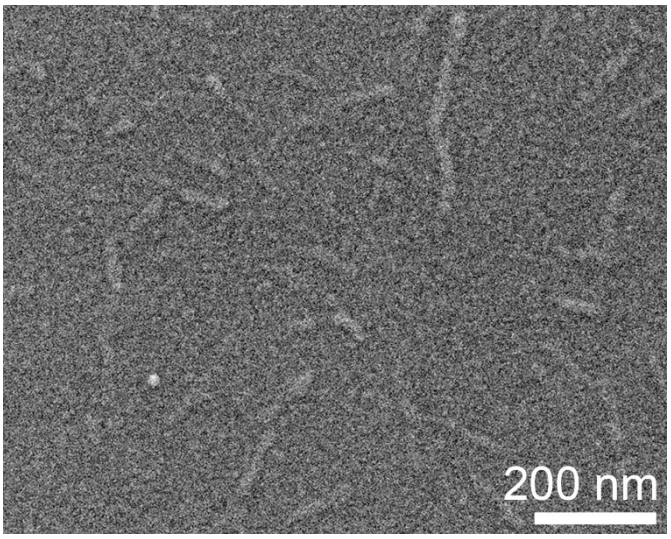


Figure S1. SEM image of a Co-P3HT thin film with nanorods obtained at α = 0.32.

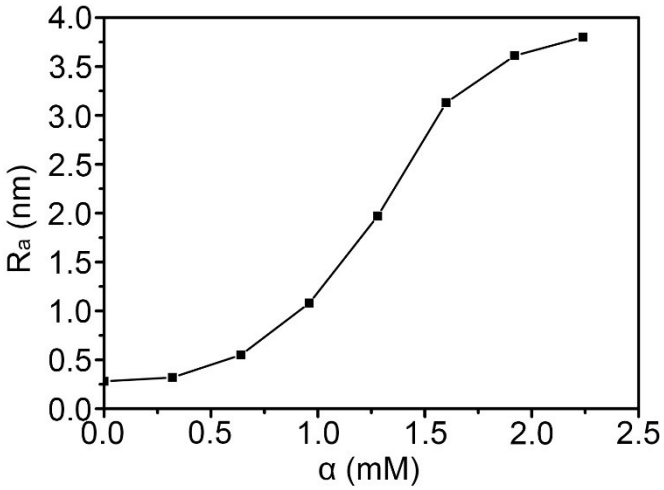


Figure S2. Average surface roughness (R_a) of P3HT thin films obtained at α = 0, 0.32, 0.64, 0.96, 1.28, 1.6, 1.92, and 2.24.

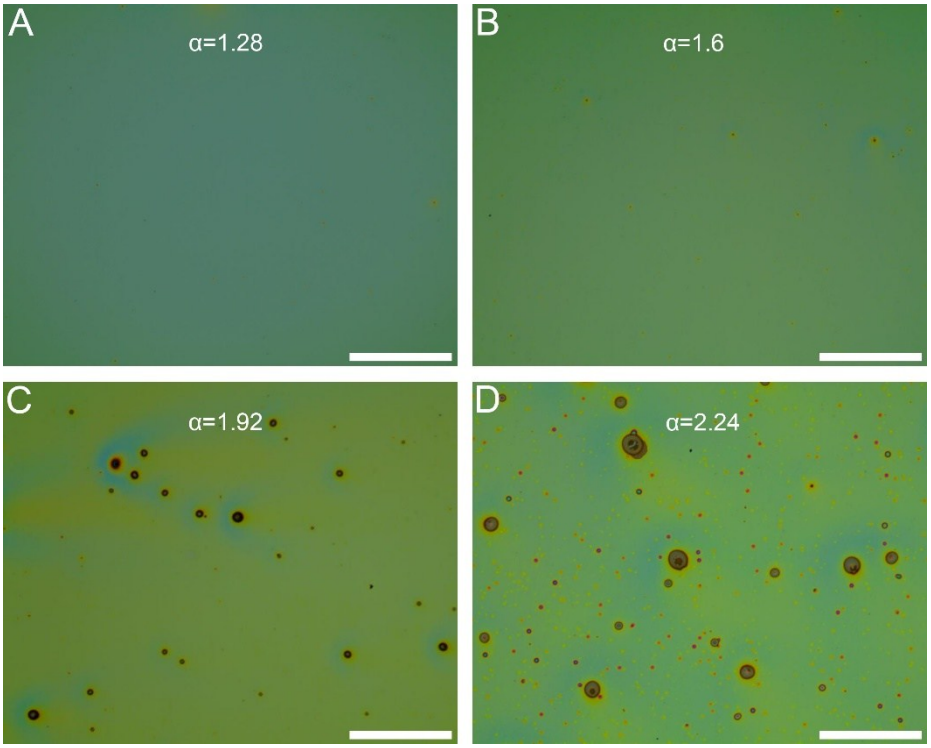


Figure S3. Optical microscope (OM) images of Co-P3HT thin films obtained at α = 1.28, 1.6, 1.92, and 2.24. The scale bars are 100 μm.

Figure S4. Photograph of the Co(II)-TFSI-doped P3HT solution (left) and the Co(III)-TFSI-doped P3HT solution (right). They were kept at 60 °C for 24 h.

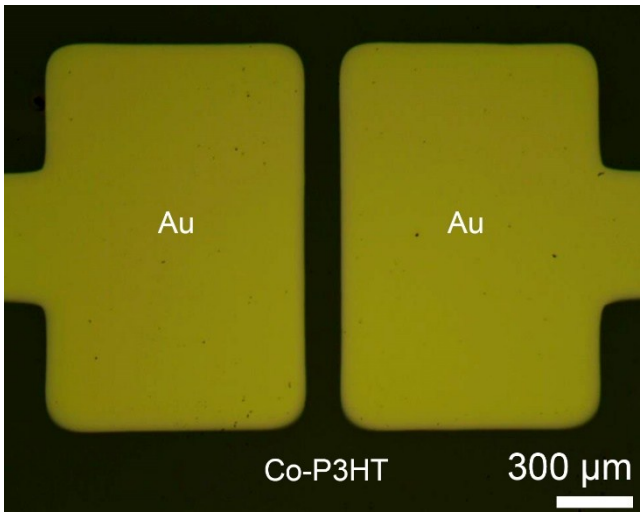


Figure S5. OM image of the 2-terminal device used to measure the conductivity of Co-P3HT thin films.

Table S1. Time-resolved photoluminescence (TRPL) parameters fitted using a bi-exponential decay function.[a)]

Co(II)-TFSI [α, mM]	a_1	τ_1 [ns]	a_2	τ_2 [ns]	τ_{avg}[b)] [ns]
0	0.503	2.671	0.497	37.1	19.78
0.32	0.526	2.407	0.474	25.72	13.45
0.64	0.511	2.485	0.489	20.25	11.17
0.96	0.554	2.101	0.446	17.16	8.82
1.28	0.680	1.896	0.320	17.71	6.95
1.6	0.511	1.482	0.303	10.53	3.94
1.92	0.670	2.008	0.330	19.11	7.65
2.24	0.665	2.176	0.345	19.796	8.27

a)Fit function = $a_1 e^{-t/\tau_1} + a_2 e^{-t/\tau_2}$

b) $\tau_{avg} = (\sum_i a_i \tau_i)/(\sum_i a_i), where \sum_i a_i = 1$

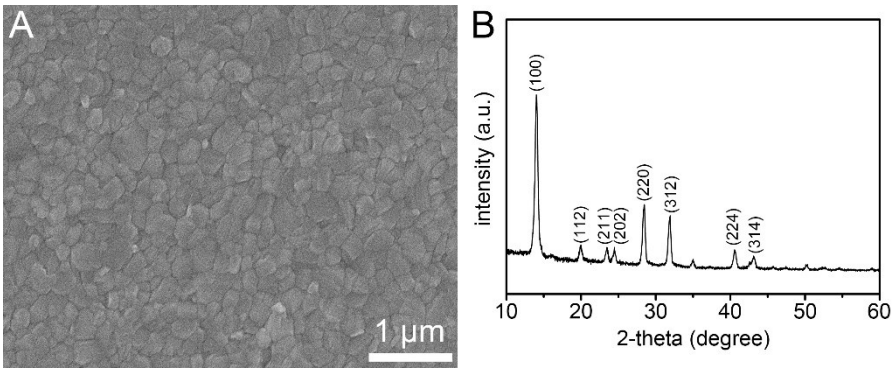


Figure S6. (A) SEM image of the $CH_3NH_3PbI_3$ thin film deposited onto the PCBM layer. (B) XRD spectrum of the $CH_3NH_3PbI_3$ thin film.

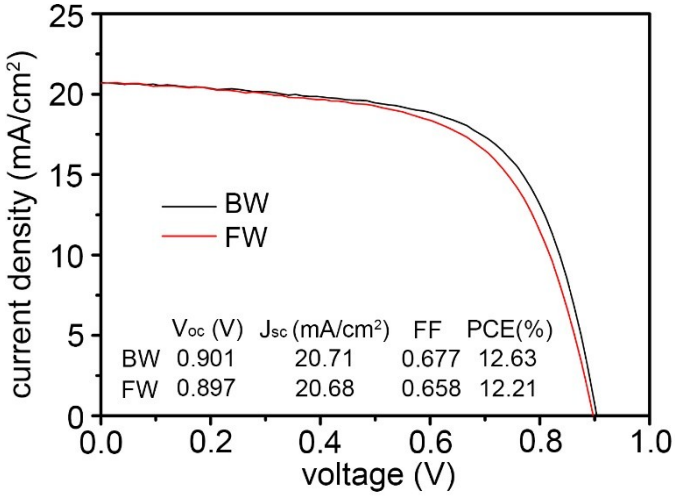


Figure S7. J-V curves of the Li-P3HT device with an active area of 0.12 cm^2 measured with a backward-forward scan for 200 ms of scan-delay time.

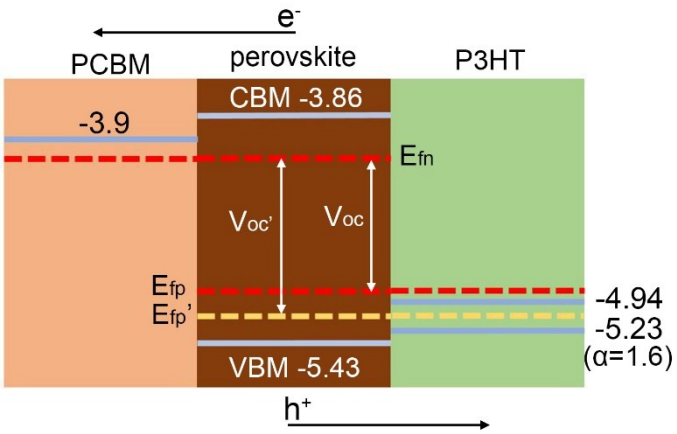


Figure S8. Energy diagram describing band alignment in the PSCs (ITO-PEIE/PCBM/Perovskite/Co-P3HT/Au). When light is transmitted to the perovskite, hole-quasi Fermi level (E_{fp}) and electron-quasi Fermi level (E_{fn}) are generated. E_{fp} is shifted to E_{fp}' in proportion with the downward shift of HOMO levels in P3HT from -4.94 eV to -5.23 eV, which produces an enhanced open-circuit voltage (V_{oc}').

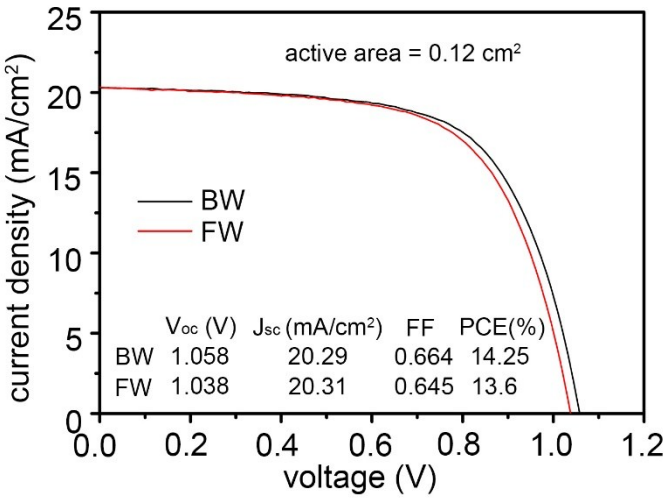


Figure S9. J-V curves of the flexible Co-P3HT device with an active area of 0.12 cm^2 measured with a backward-forward scan for 200 ms of scan-delay time.

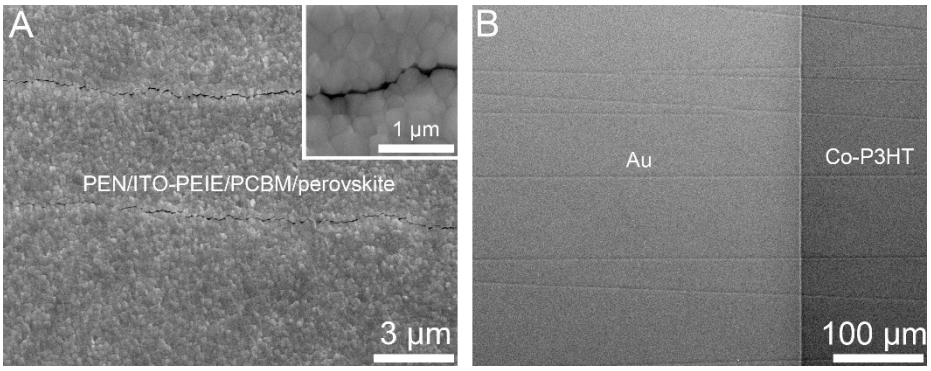


Figure S10. SEM images of (A) the perovskite on PEN/ITO-PEIE/PCBM and (B) the flexible device obtained after recovery from bending at r = 4 mm.

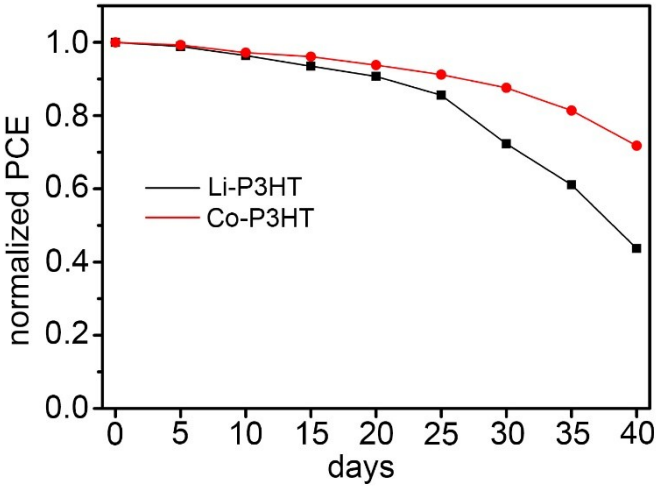
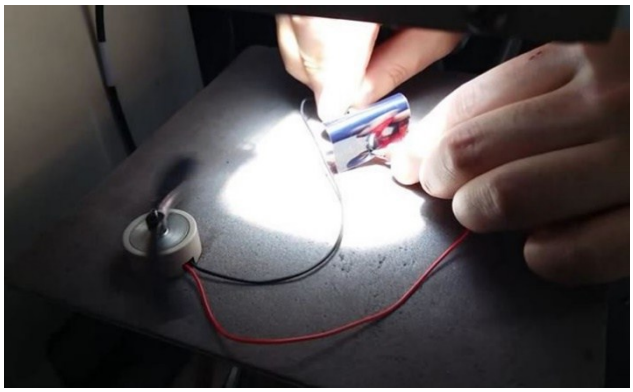


Figure S11. Normalized PCEs of the flexible Li-P3HT device and the flexible Co-P3HT device obtained under dark condition and a relative humidity of 20 % for 40 d..



Video S1. A movie showing the bending and twisting test of the flexible device incorporating Co-P3HT under 1 sun condition.