Supplementary Information

Molecular Aggregation Method Perovskite–Fullerene Bulk Hetero Structure Solar Cells

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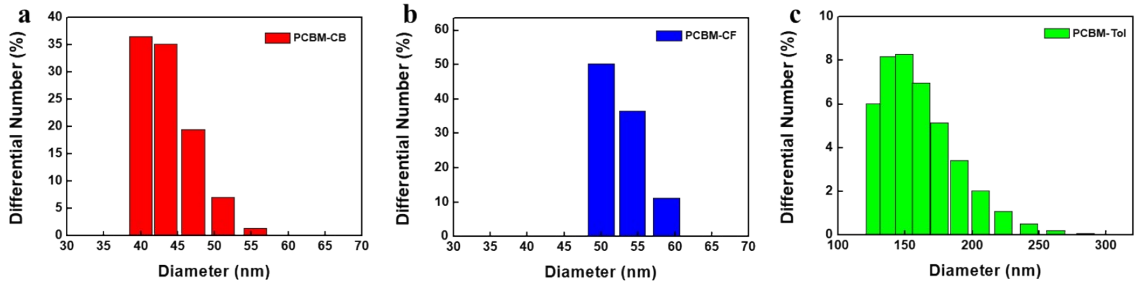

Fig. S1 Dynamic light scattering size distribution of the solvation of PCBM solutes in different solvents. (a) CB, (b) CF, and (c) Tol

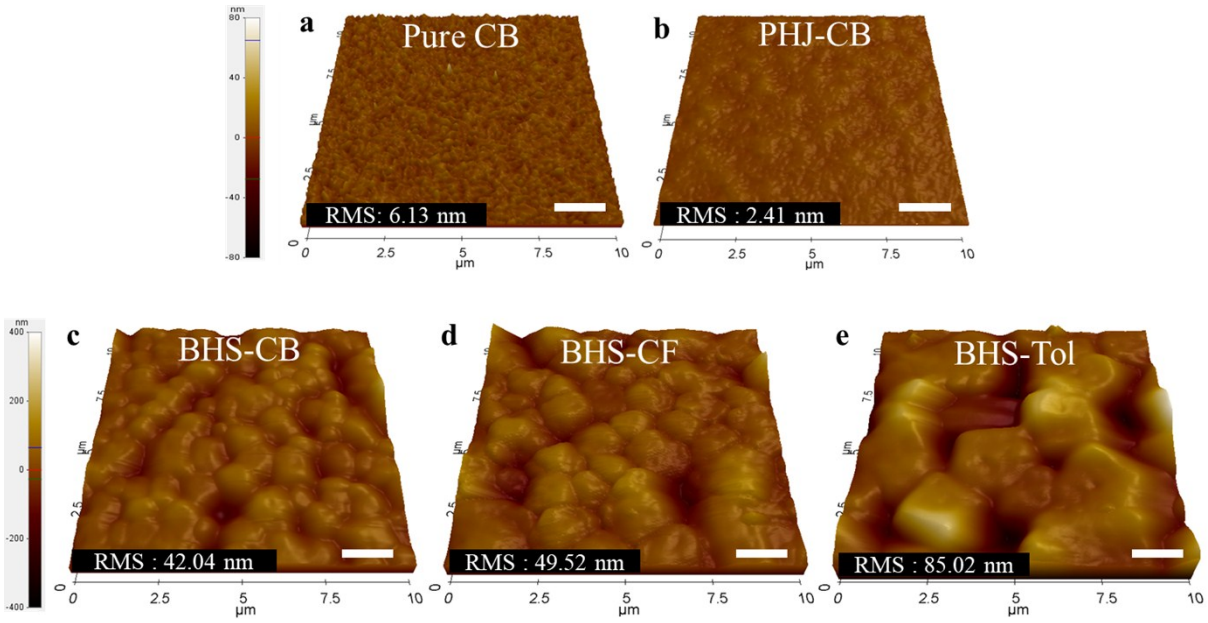

Fig. S2 AFM topographic images of perovskite films. a, perovskite film treated with pure CB anti-solvent. b, PCBM film coated on sample a (PHJ-CB). c-e, Perovskite films with Pe-PCBM BHS using CB solvent (BHS-CB) (c), CF (d), and Tol (e). Perovskite films with PHJ and Pe-PCBM BHS have sample configuration of ITO/PEDOT:PSS/perovskite/PCBM. Scale bar is 2 µm.

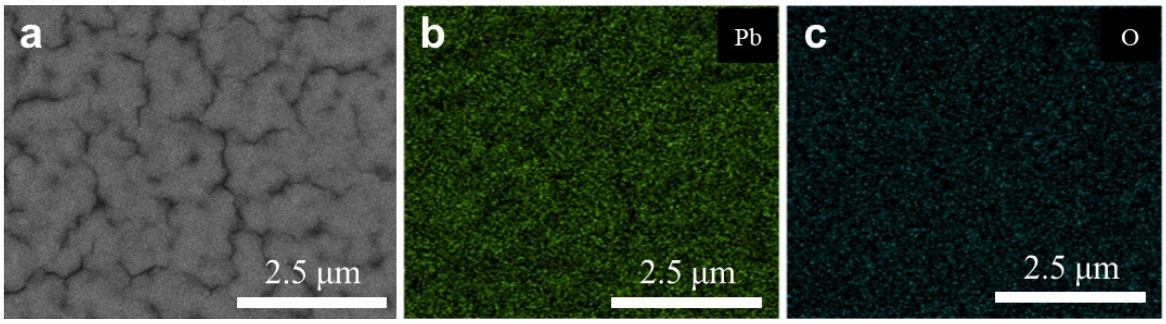

Fig. S3 SEM and EDS mapping images of PHJ film. a-c, Top-veiw SEM image (a) and EDS mapping profiles of SEM image (a) showing Pb (green, b) of perovskite and O (cyan, c) elements of PCBM, repsectively. Scare bar is 2.5 μm.

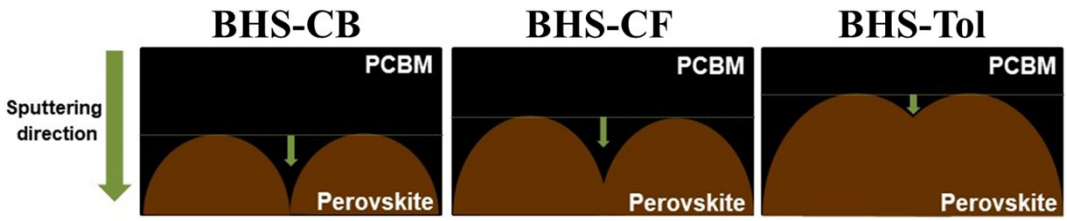

Fig. S4 Images for comparing reduction rate for amount of PCBM in Pe-PCBM BHS films.

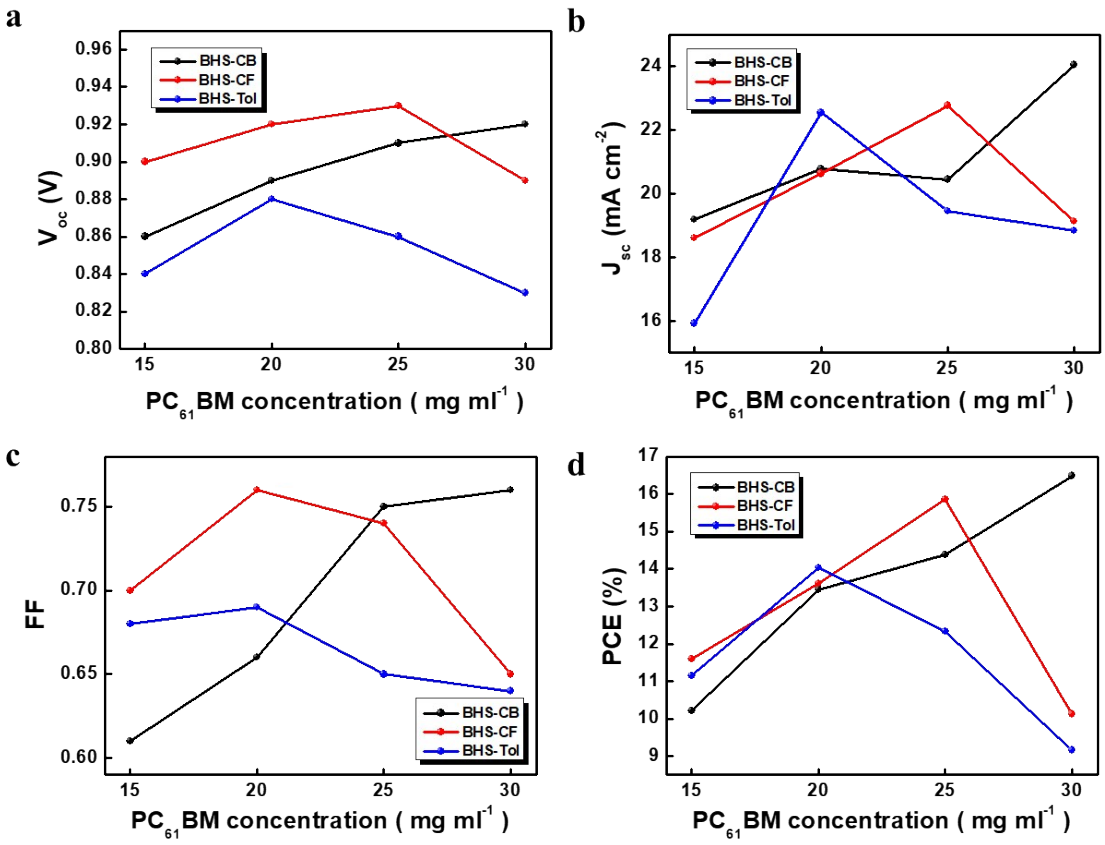

Fig. S5 Optimization of the devices with Pe-PCBM BHS as a function of PCBM thickness. a-d, V_{OC} (a), J_{SC} (b), FF (c), and PCE (d).

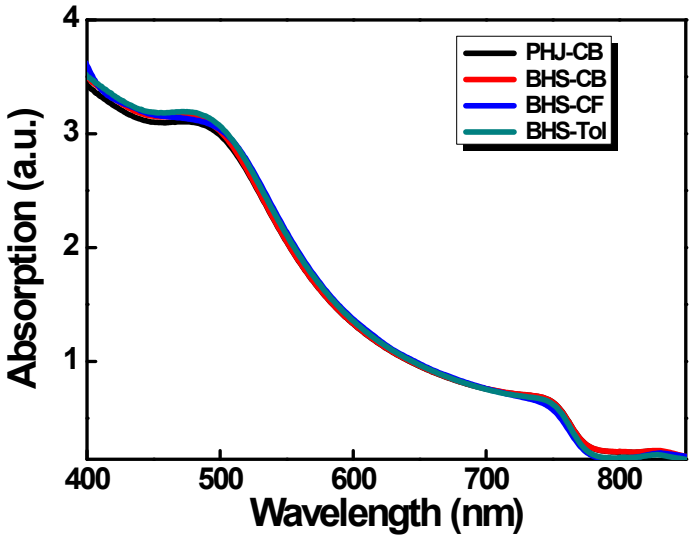

Fig. S6 UV-vis absorption of perovskite films with PHJ and Pe-PCBM BHS. Absorption spectra of perovskite/PCBM (PHJ) and perovskite:PCBM (BHS) films.

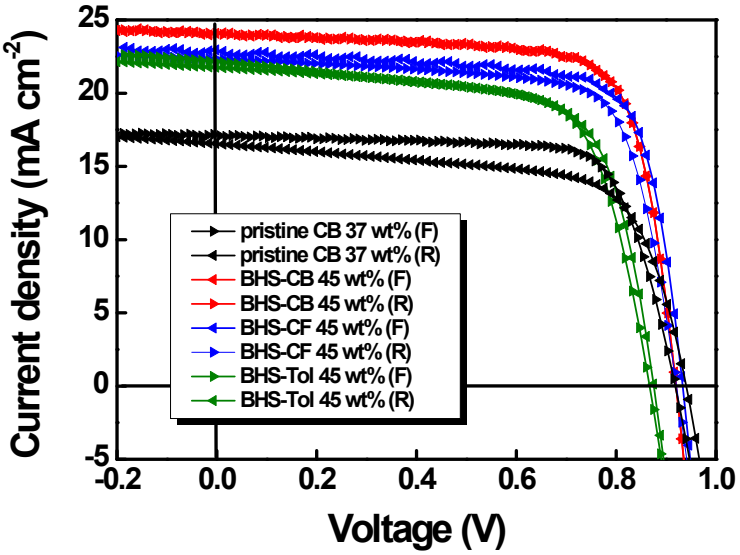

Fig. S7 Hysteresis of devices with PHJ and Pe-PCBM BHS using different solvents. Scan rate for forward and reverse directions is 30 mV s^{-1}.

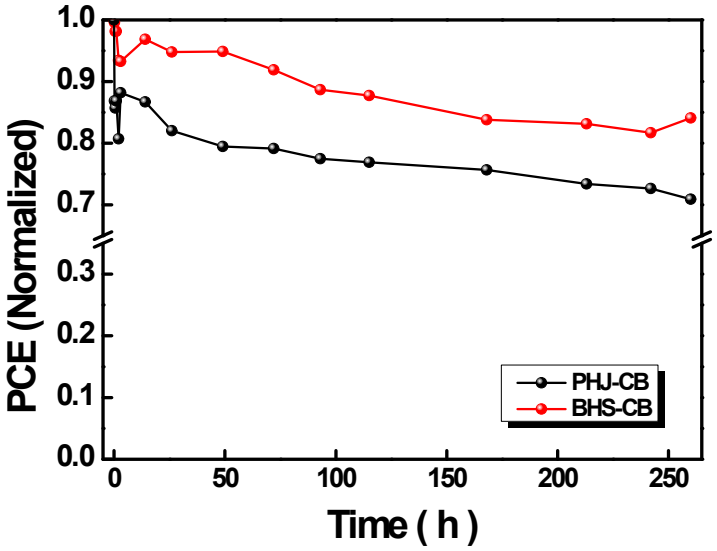

Fig. S8 Comparison of device stability between PHJ and Pe-PCBM BHS.

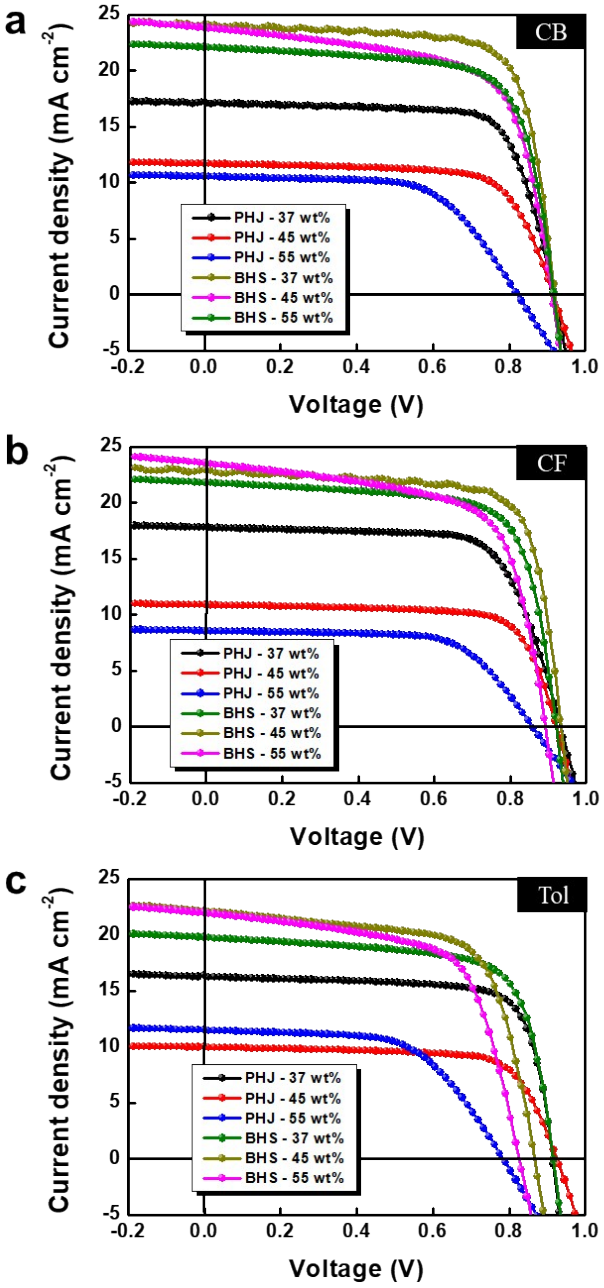

Fig. S9 *J-V* curves of the devices as a function of structure, solvent and perovskite thickness. a-c, *J-V* curves of the devices with PHJ and Pe-PCBM BHS using CB (a), CF (b), and Tol (c).

Table S1 Comparison of our works with previous reports published in journal with high quality.

Reference	Perovskite -PCBM structure	Perovskite materials	Device structure	PCE (%)	J_{sc} (mA cm^{-2})	V_{OC} (V)	FF
Wu's group (Nat. Photon., 2016)	BHJ	$CH_3NH_3PbI_3$	ITO / PEDOT:PSS / PHJ[a] / Ca / Al	11.4	16.2	0.95	0.73
			ITO / PEDOT:PSS / BHJ / Ca / Al	12.1	16.0	0.96	0.79
			ITO / PEDOT:PSS / BHJ / $PC_{71}BM$ / Ca / Al	16.0	20.2	0.97	0.82
Han's group (Nat. Energy, 2016)	GHJ	$FA_{0.85}MA_{0.15}Pb(I_{0.85}Br_{0.15})_3$	FTO / NiO_x / PHJ / Nb-TiO_x / Al	11.31	17.90	0.93	0.68
			FTO / NiO_x / GHJ / Al		Not reported		
			FTO / NiO_x / GHJ / PCBM / Nb-TiO_x / Al	18.21	21.95	1.08	0.80
		$CH_3NH_3PbI_3$	FTO / NiO_x / PHJ / Nb-TiO_x / Al	16.1	20.01	1.03	0.78
			FTO / NiO_x / GHJ / Al		Not reported		
			FTO / NiO_x / GHJ / PCBM / Nb-TiO_x / Al	16.9	20.09	1.04	0.78
Our work	BHS	$CH_3NH_3PbI_3$	ITO / PEDOT:PSS / PHJ / Al	11.65	17.09	0.92	0.74
			ITO / PEDOT:PSS / BHS / Al	16.49	24.05	0.92	0.75
			ITO / PEDOT:PSS / BHS / PCBM / Al	18.08	24.15	0.97	0.77

[a] This PHJ structure consists of $CH_3NH_3PbI_3$ and $PC_{71}BM$.

Table S2 The photovoltaic parameters for hysteresis of the devices with PHJ and Pe-PCBM BHS using different solvents.

Device type	Scan	V_{oc} (V)	J_{sc} (mA cm^{-2})	FF	PCE (%)
PHJ (CB)	Forward	0.91	17.98	0.72	11.78
	Reverse	0.91	17.40	0.71	11.32
BHS (CB)	Forward	0.92	24.05	0.75	16.49
	Reverse	0.92	24.09	0.74	16.36
BHS (CF)	Forward	0.93	22.77	0.75	15.85
	Reverse	0.92	22.32	0.74	15.22
BHS (Tol)	Forward	0.88	22.54	0.69	13.81
	Reverse	0.87	21.75	0.69	13.14

Table S3 Photovoltaic parameters of the devices as a function of structure, solvent and perovskite thickness.

Device type	Perovskite concentration (wt %)	V_{oc} (V)	J_{sc} (mA cm^{-2})	FF	PCE (%)
PHJ (CB)	37	0.92 (± 0.01)	17.09 (± 0.48)	0.74 (± 0.01)	11.65 (± 0.43)
	45	0.92 (± 0.01)	11.68 (± 0.34)	0.71 (± 0.02)	7.60 (± 0.31)
	55	0.82 (± 0.02)	10.54 (± 0.39)	0.63 (± 0.02)	5.42 (± 0.39)
PHJ (CF)	37	0.93 (± 0.01)	17.78 (± 0.41)	0.71 (± 0.02)	11.78 (± 0.32)
	45	0.93 (± 0.01)	10.87 (± 0.38)	0.72 (± 0.02)	7.39 (± 0.21)
	55	0.85 (± 0.03)	8.55 (± 0.34)	0.65 (± 0.03)	4.82 (± 0.46)
PHJ (Tol)	37	0.92 (± 0.01)	16.26 (± 0.37)	0.76 (± 0.02)	11.29 (± 0.33)
	45	0.92 (± 0.02)	9.95 (± 0.41)	0.72 (± 0.01)	6.65 (± 0.25)
	55	0.78 (± 0.02)	11.48 (± 0.43)	0.60 (± 0.04)	5.34 (± 0.39)
BHS (CB)	37	0.92 (± 0.01)	21.87 (± 0.35)	0.71 (± 0.01)	14.46 (± 0.36)
	45	0.92 (± 0.01)	24.05 (± 0.42)	0.75 (± 0.02)	16.49 (± 0.31)
	55	0.91 (± 0.01)	23.35 (± 0.48)	0.64 (± 0.01)	14.02 (± 0.43)
BHS (CF)	37	0.92 (± 0.01)	21.89 (± 0.41)	0.70 (± 0.02)	14.10 (± 0.32)
	45	0.93 (± 0.01)	22.77 (± 0.36)	0.75 (± 0.02)	15.85 (± 0.25)
	55	0.90 (± 0.02)	22.97 (± 0.39)	0.64 (± 0.01)	13.33 (± 0.39)
BHS (Tol)	37	0.91 (± 0.01)	19.56 (± 0.32)	0.70 (± 0.02)	12.45 (± 0.33)
	45	0.88 (± 0.03)	22.54 (± 0.38)	0.69 (± 0.03)	13.81 (± 0.42)
	55	0.82 (± 0.01)	21.55 (± 0.42)	0.63 (± 0.02)	11.60 (± 0.31)