Supporting information

Synthesis, Characterization and Photovoltaic Properties of Poly(cyclopentadithiophene-alt-isoindigo)

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The NMR spectra of synthesized monomers and polymers are illustrated below from **Figure S1** to **S8**.

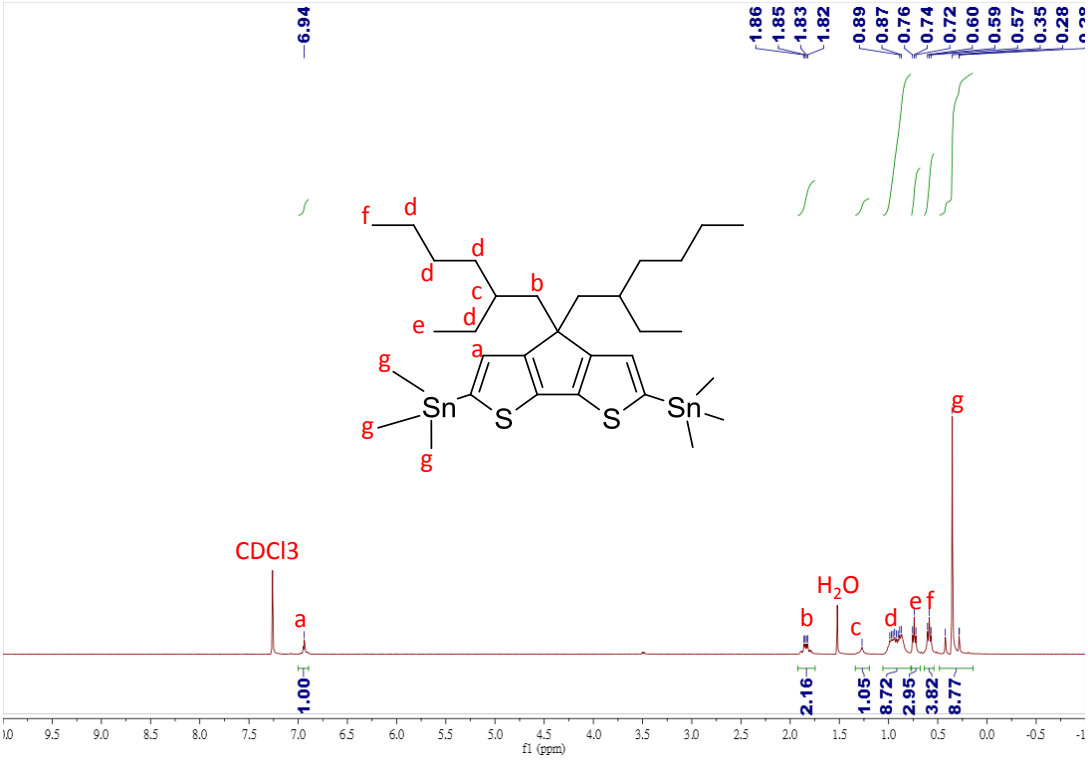

Figure S1. [1]H NMR of 6a.

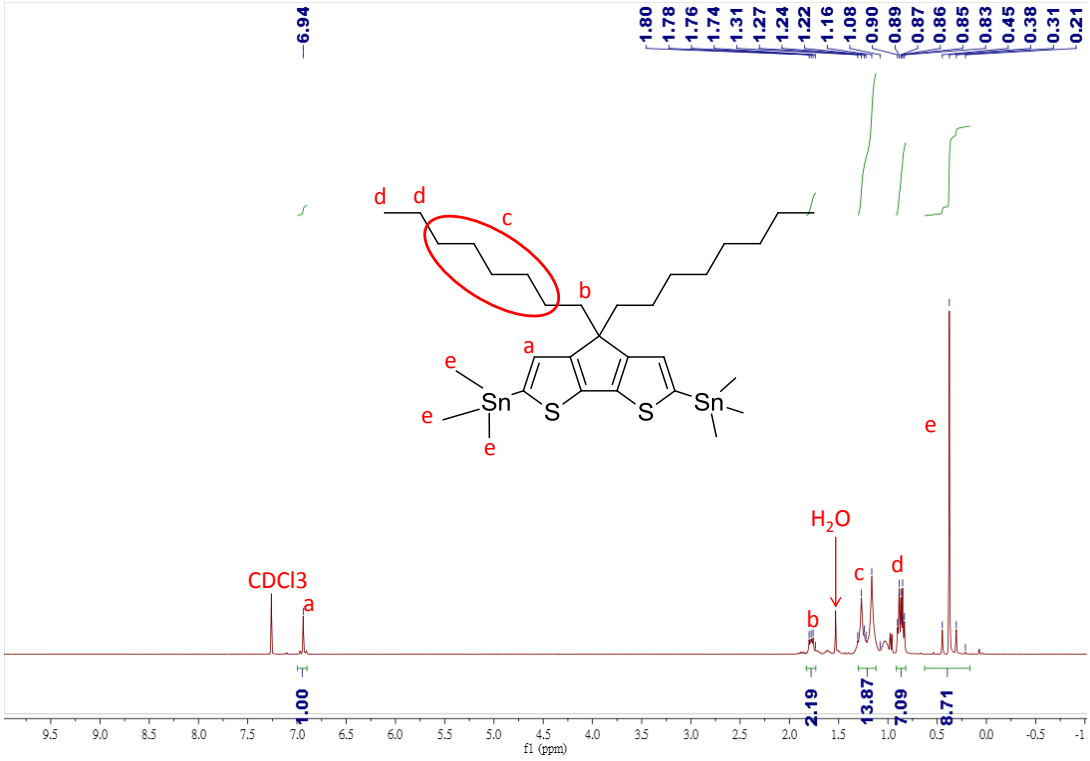

Figure S2. ^{1}H NMR of 6b.

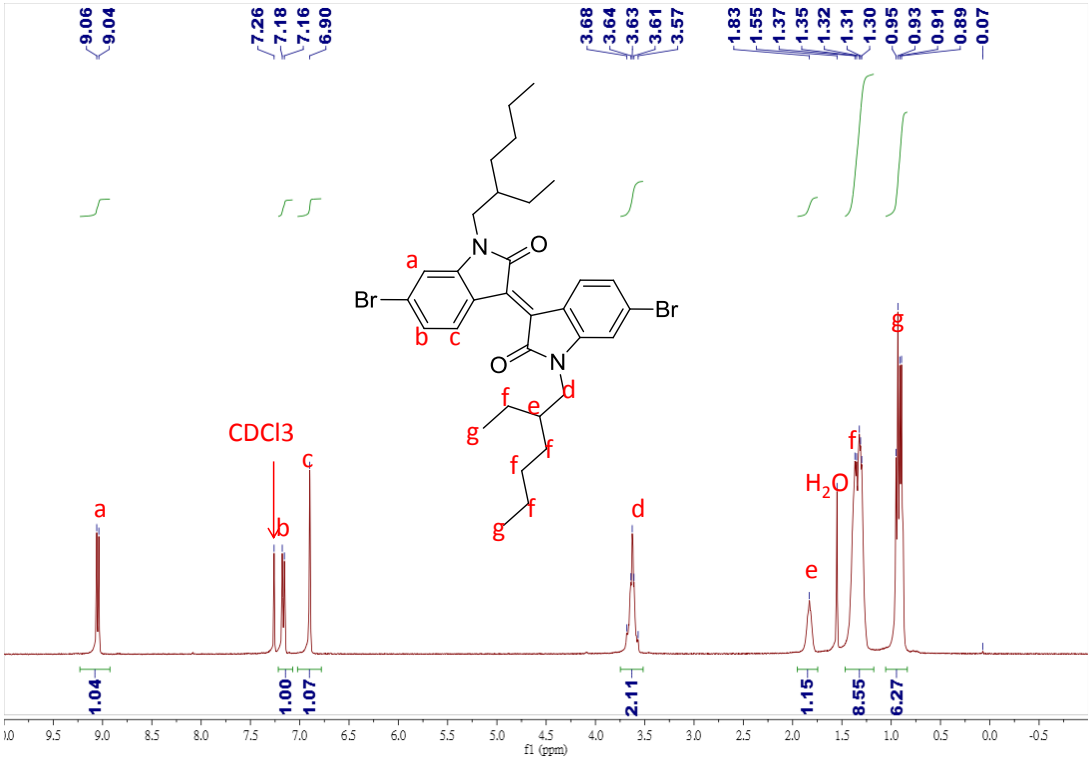

Figure S3. ^{1}H NMR of 8a.

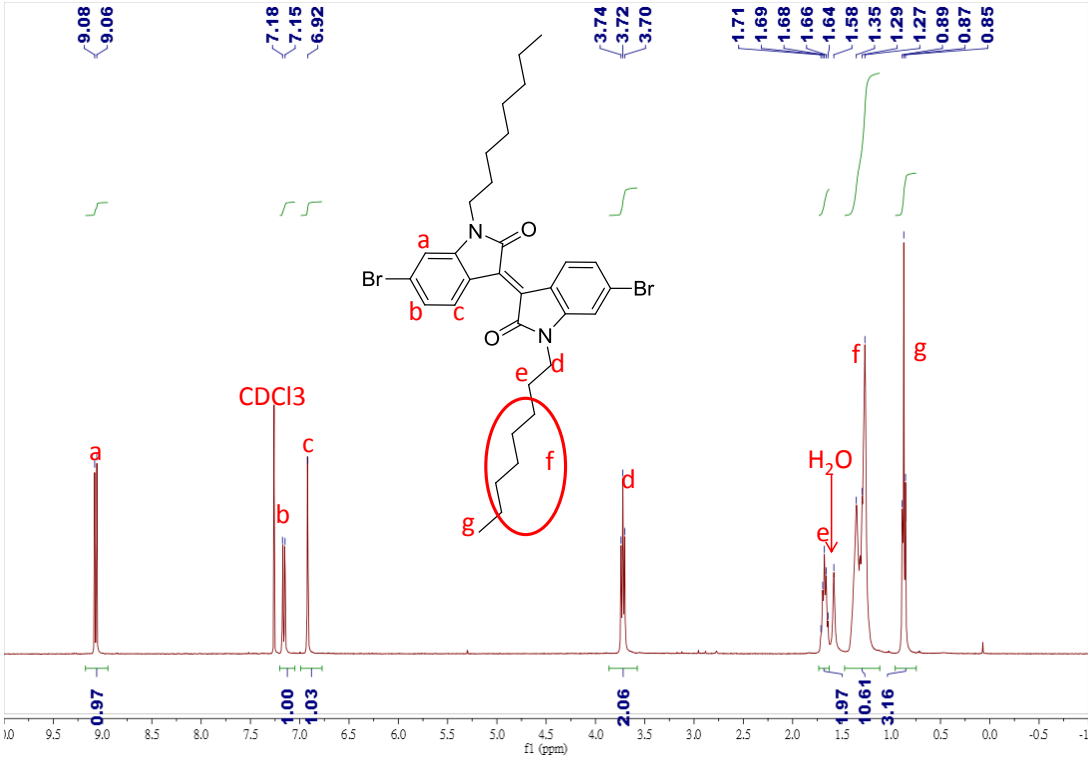

Figure S4. ^{1}H NMR of 8b.

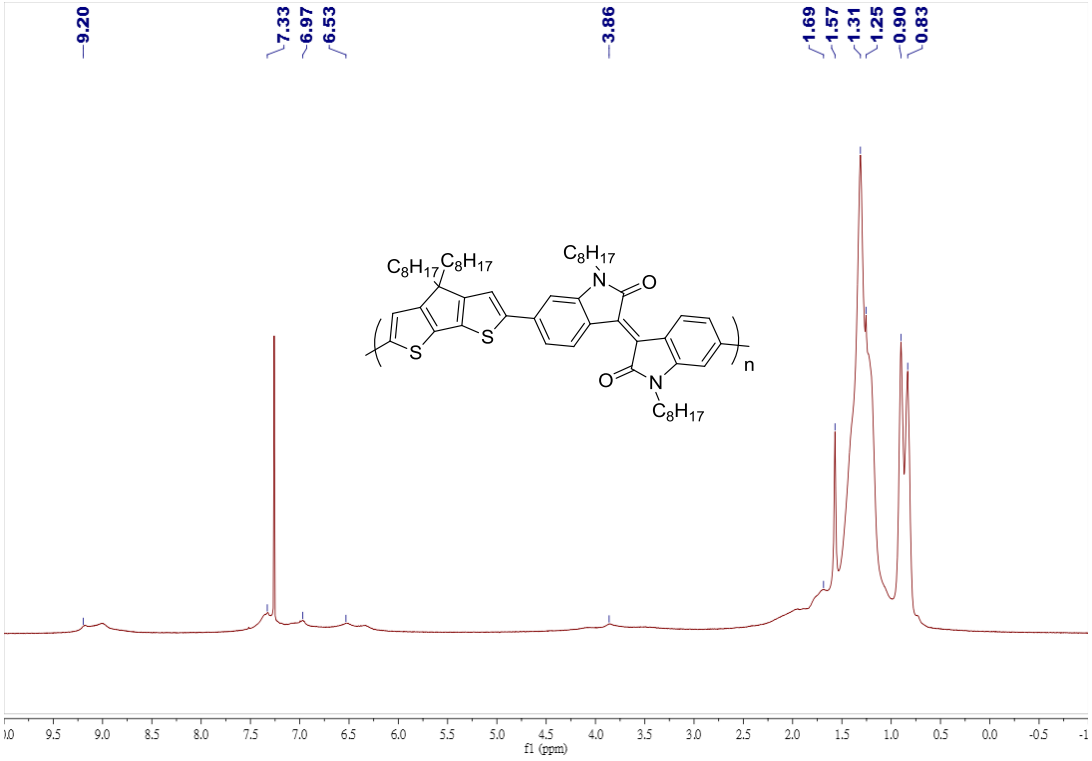

Figure S5. ^{1}H NMR of PC8I8.

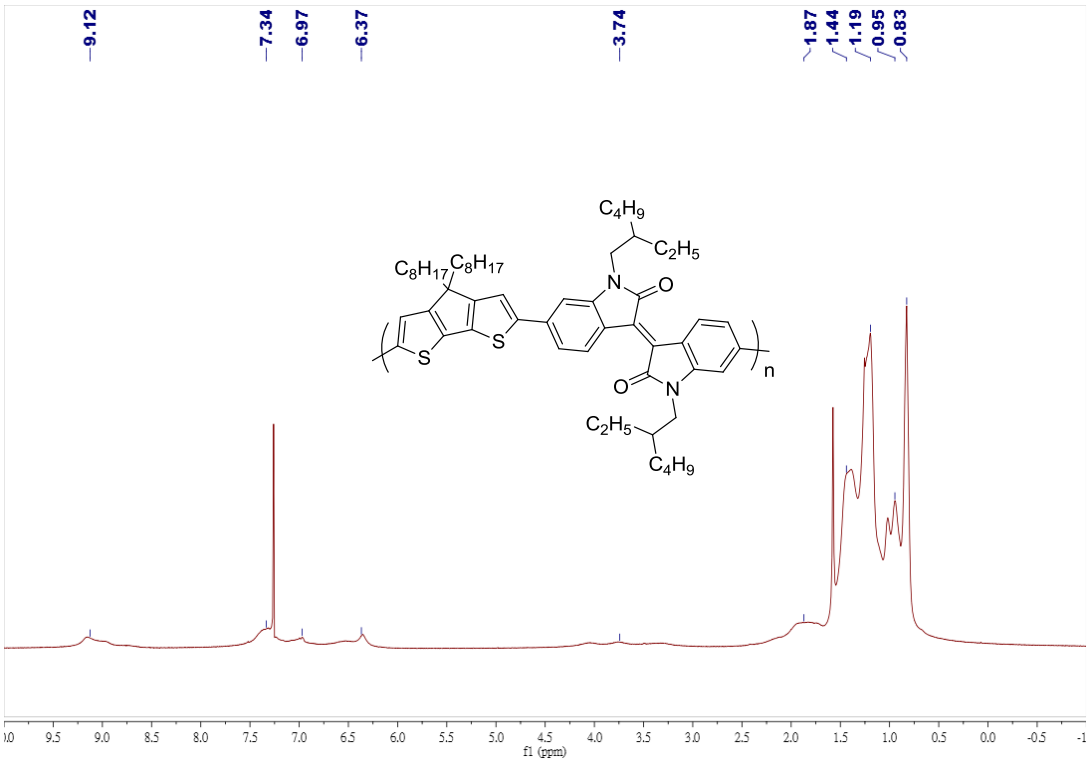

Figure S6. ^{1}H NMR of PC8Ie

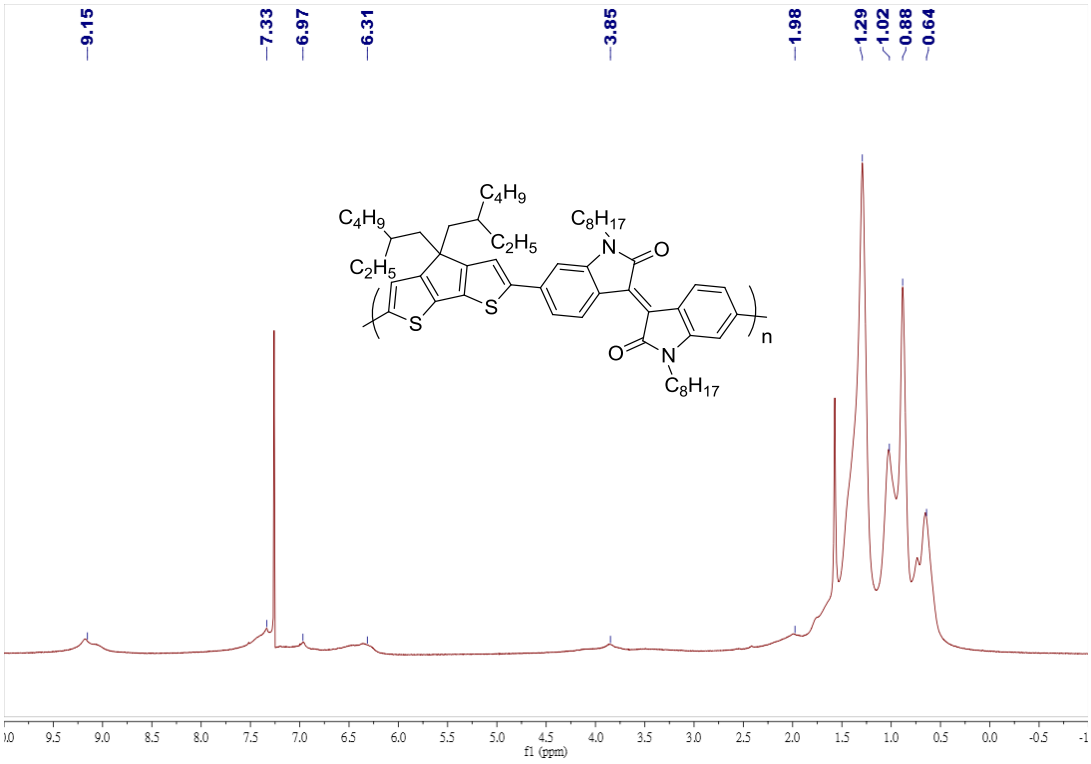

Figure S7. ^{1}H NMR of PCeI8

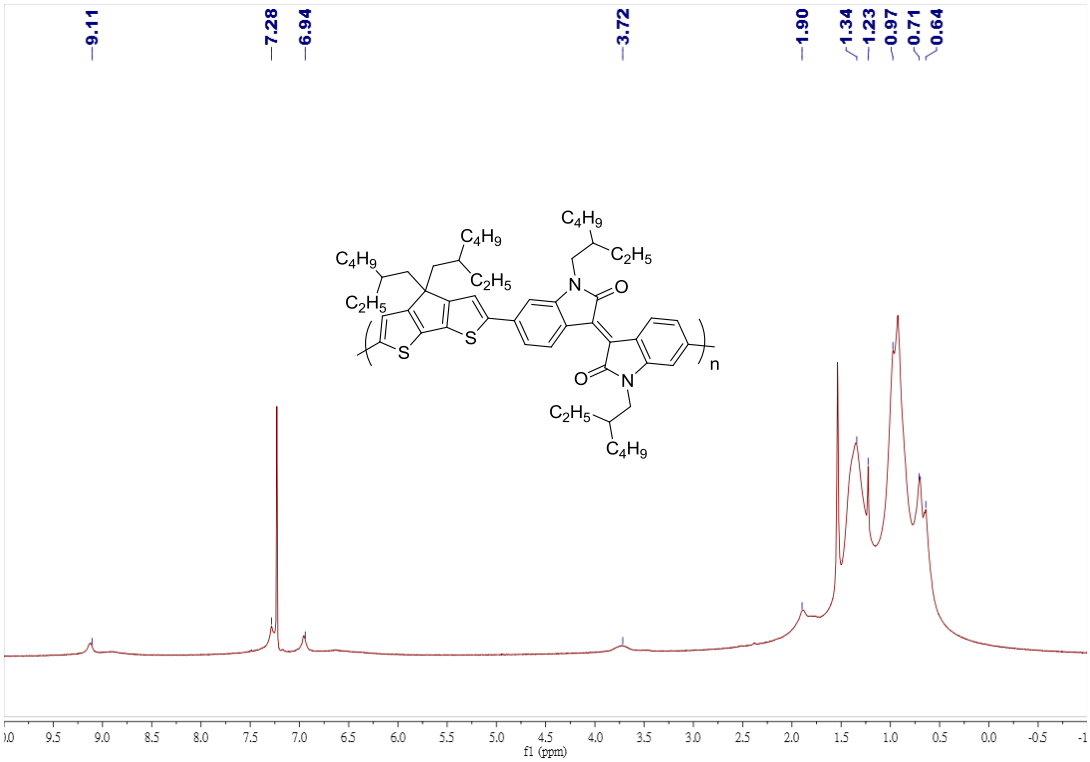

Figure S8. ^{1}H NMR of PCeIe

The AFM profiles of the isoindigo polymer/PC61BM thin films are shown below from **Figures S9 to S12**. We can find that the difference of domain size between mixture ratio of 1:1 and 1:2 is discernible, especially in the cases processed by chlorobenzene (CB). The size of aggregation is affected by the side chain type of copolymer but may also be influence by the molecular weight of the copolymer.

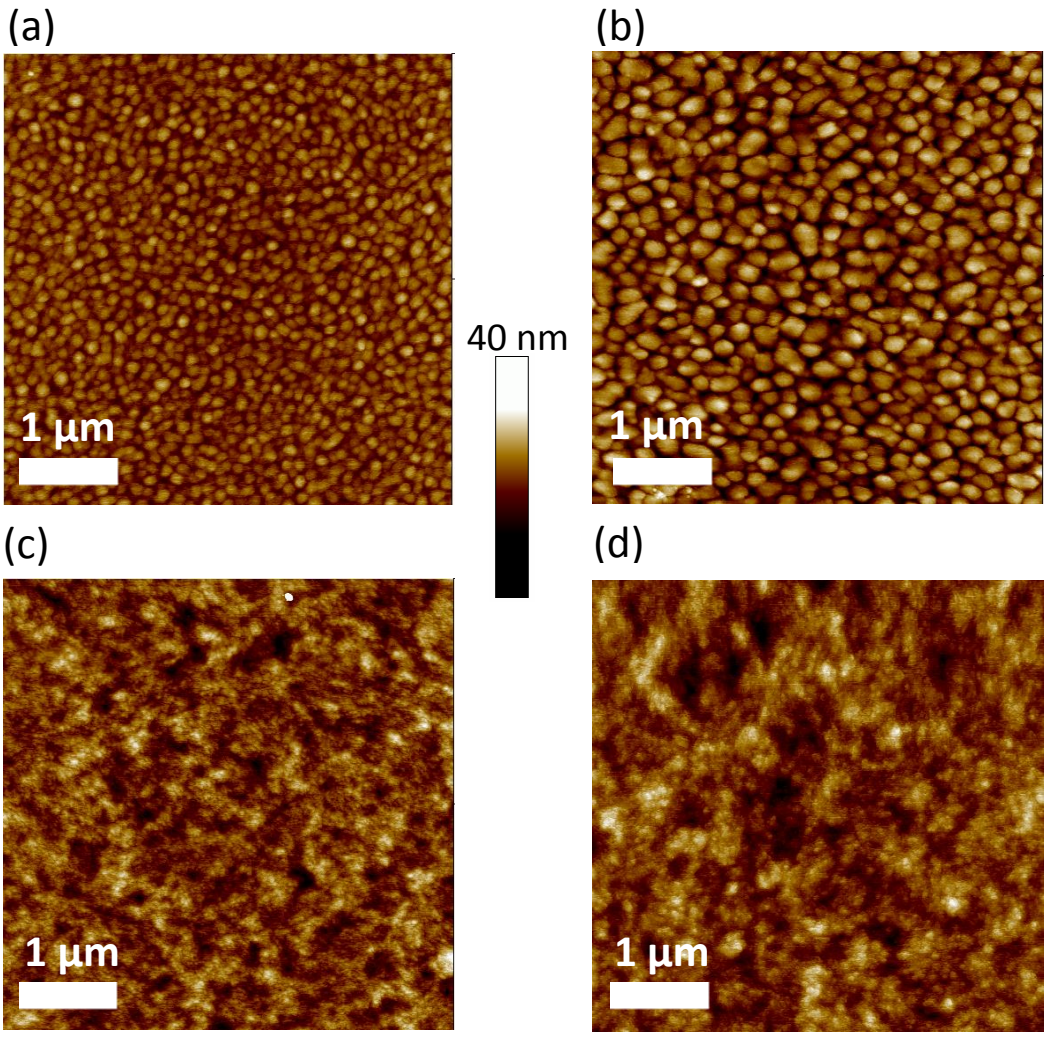

Figure S9. 2D tapping mode AFM surface scans of PC8I8 (Mn= 25k) /PC61BM at a ratio of (a) 1:1 and (b) 1:2 processed with pristine CB. (c) 1:1 (d) 1:2 processed by CB + 3% DIO.

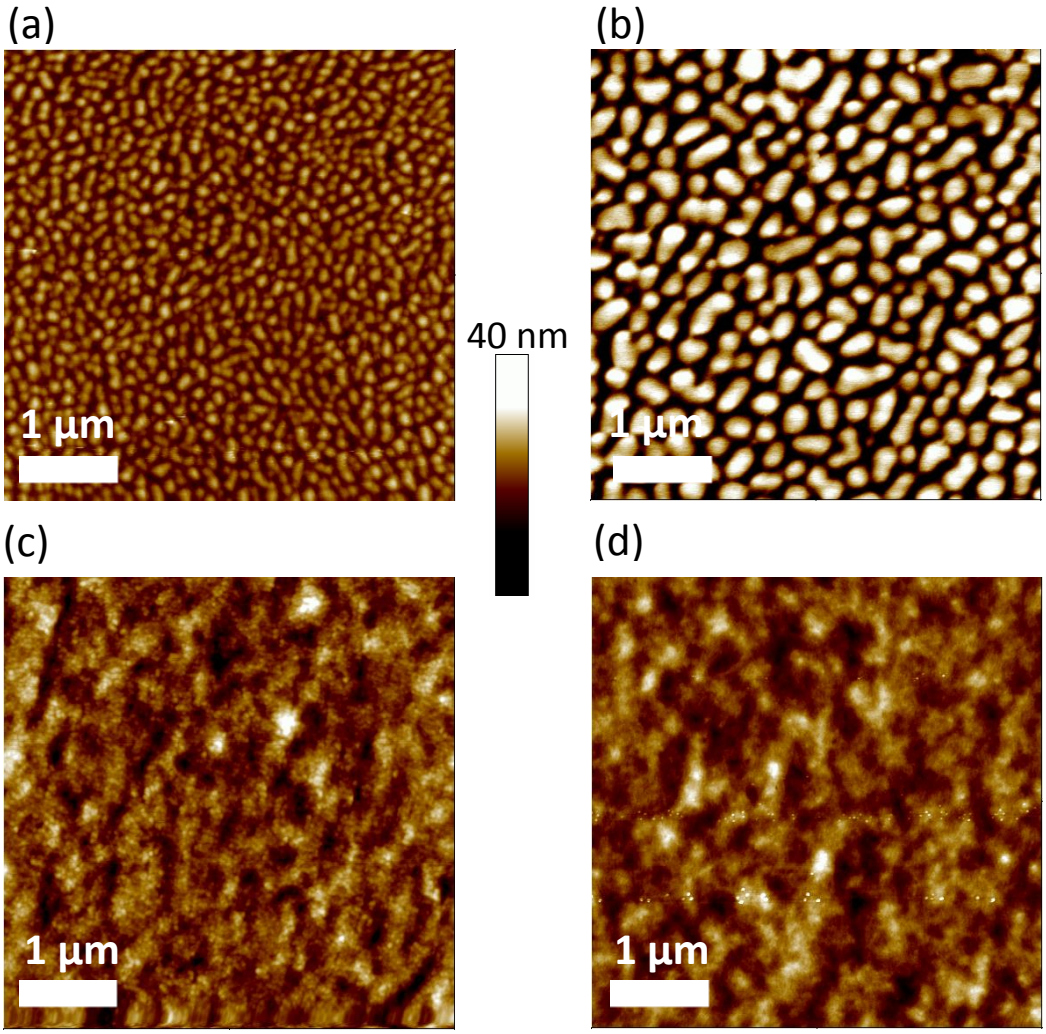

Figure S10. 2D tapping mode AFM surface scans of PC8Ie (Mn= 20k) /PC61BM at a ratio of (a) 1:1 and (b) 1:2 processed with pristine CB. (c) 1:1 (d) 1:2 processed by CB + 3% DIO.

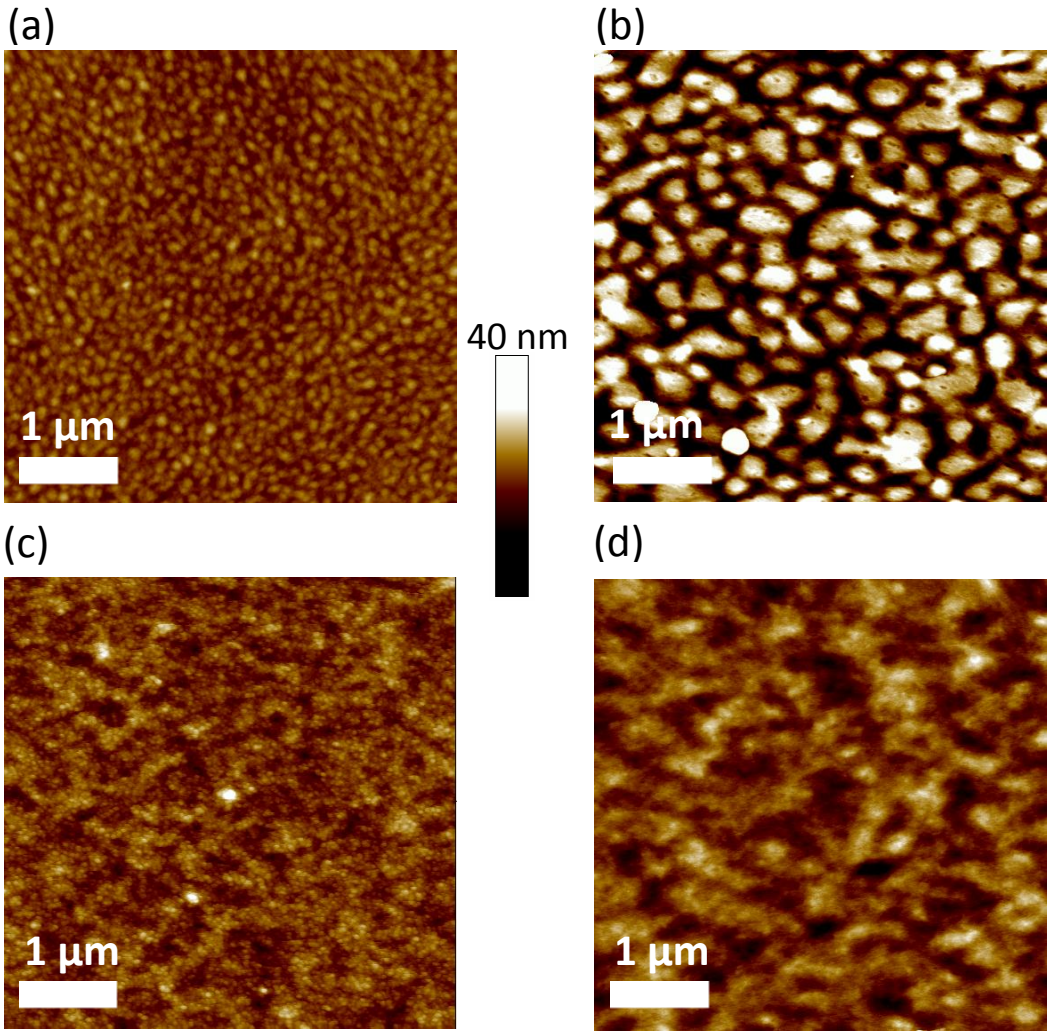

Figure S11. 2D tapping mode AFM surface scans of PCeI8 (Mn= 36k) /PC61BM at a ratio of (a) 1:1 and (b) 1:2 processed with pristine CB. (c) 1:1 (d) 1:2 processed by CB + 3% DIO.

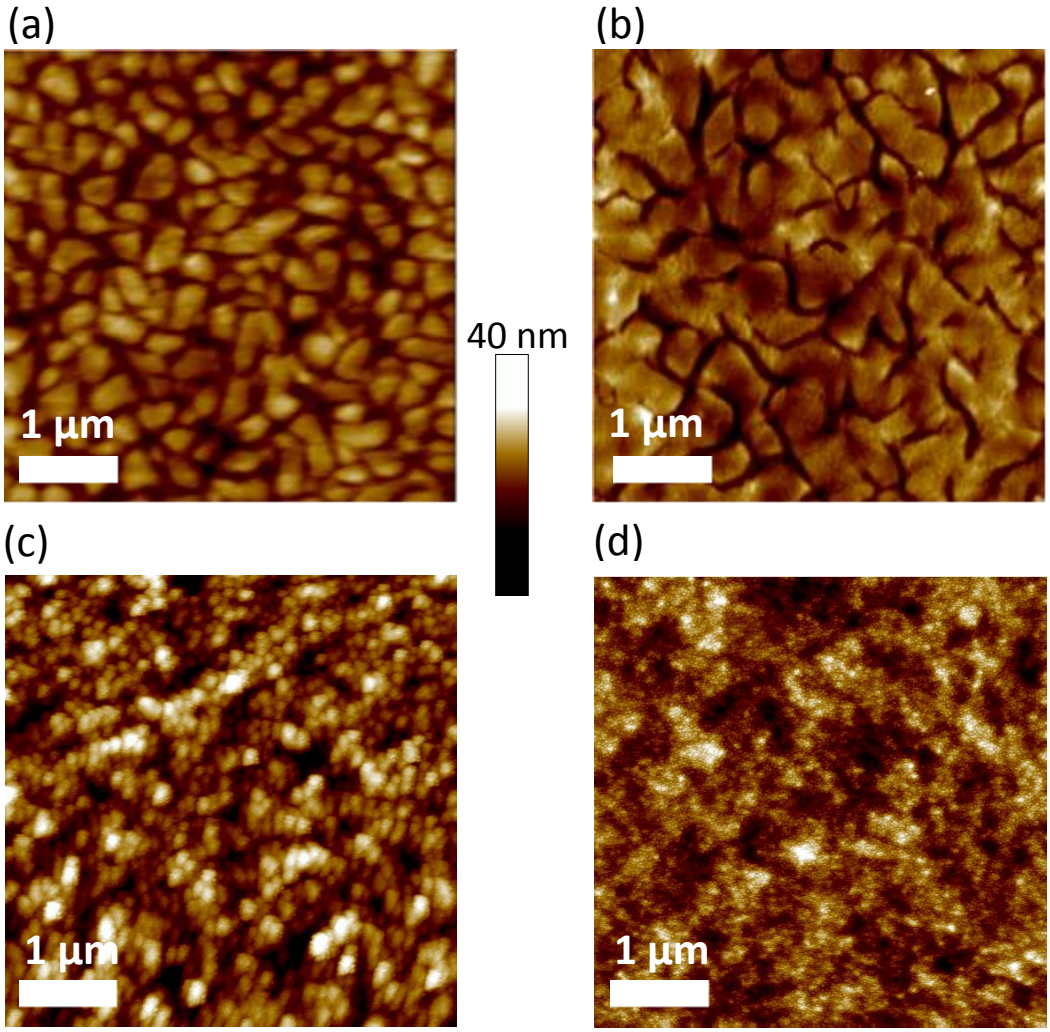

Figure S12. 2D tapping mode AFM surface scans of PCeIe (Mn= 45k) /PC61BM at a ratio of (a) 1:1 and (b) 1:2 processed with pristine CB. (c) 1:1 (d) 1:2 processed by CB + 3% DIO.

Table S1. Summary of device performance for devices fabricated from (a) PC8I8/PC61BM and PC8Ie/PC61BM with 3% DIO respectively, (b) PCeI8/PC61BM and PCeIe/PC61BM with 3% DIO respectively.

(a)

	PC8I8				**PC8Ie**			
Mixture ratios	Voc (V)	Jsc (mA/cm^2)	FF (%)	PCE (%)	Voc (V)	Jsc (mA/cm^2)	FF (%)	PCE (%)
1:0.5	0.69	4.0	35.5	1.0	0.72	4.1	43.0	1.3
1:1	0.69	8.3	39.9	2.2	0.73	8.2	44.5	**2.7**
1:1.5	0.66	9.3	45.0	2.7	0.73	7.9	44.2	2.5
1:2	0.66	10.1	50.2	**3.3**	0.72	7.2	43.6	2.3
1:3	0.66	7.5	39.0	1.9	0.72	5.7	35.3	1.5

(b)

	PCeI8				**PCeIe**			
Mixture ratios	Voc (V)	Jsc (mA/cm^2)	FF (%)	PCE (%)	Voc (V)	Jsc (mA/cm^2)	FF (%)	PCE (%)
1:0.5	0.76	6.6	42.7	2.1	0.81	6.9	42.8	2.7
1:1	0.76	9.6	40.0	2.9	0.80	10.1	45.7	3.4
1:1.5	0.78	9.7	44.9	3.4	0.80	11.6	43.0	**4.0**
1:2	0.79	9.7	46.9	**3.6**	0.78	9.5	38.7	2.9
1:3	0.72	5.4	35.8	1.4	0.76	9.0	36.0	2.5