Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Three-Dimensional Directed Assembly of Organic Charge-Transfer Heterostructure

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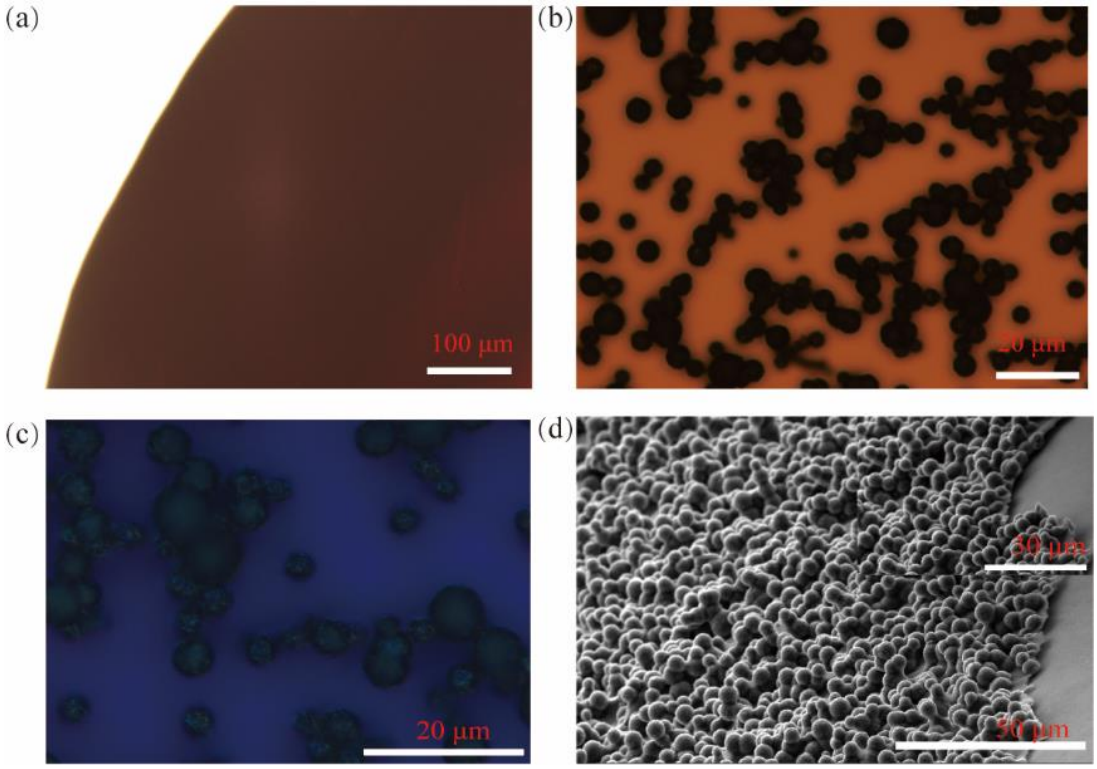

Figure S1. Optical microscopy images of (a) uniform film of the polythiophene-C_{60} solution, (b) high magnification image, (c) low magnification image, (d) side-view scanning electron microscope (SEM) images of polythiophene-C_{60} formed at 313K under 30 μL ACN atmosphere and the inset was high magnification image.

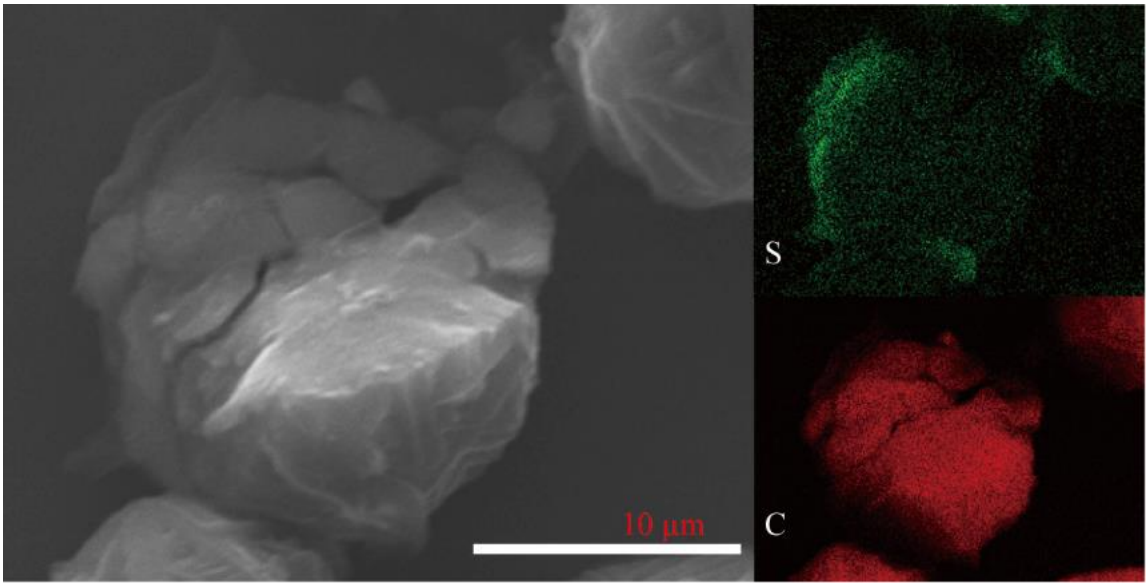

Figure S2. SEM image and the carbon and sulfur elemental mapping from energy-dispersive X-ray spectroscopy of polythiophene-C_{60} formed at 313K under 30 µL ACN atmosphere.

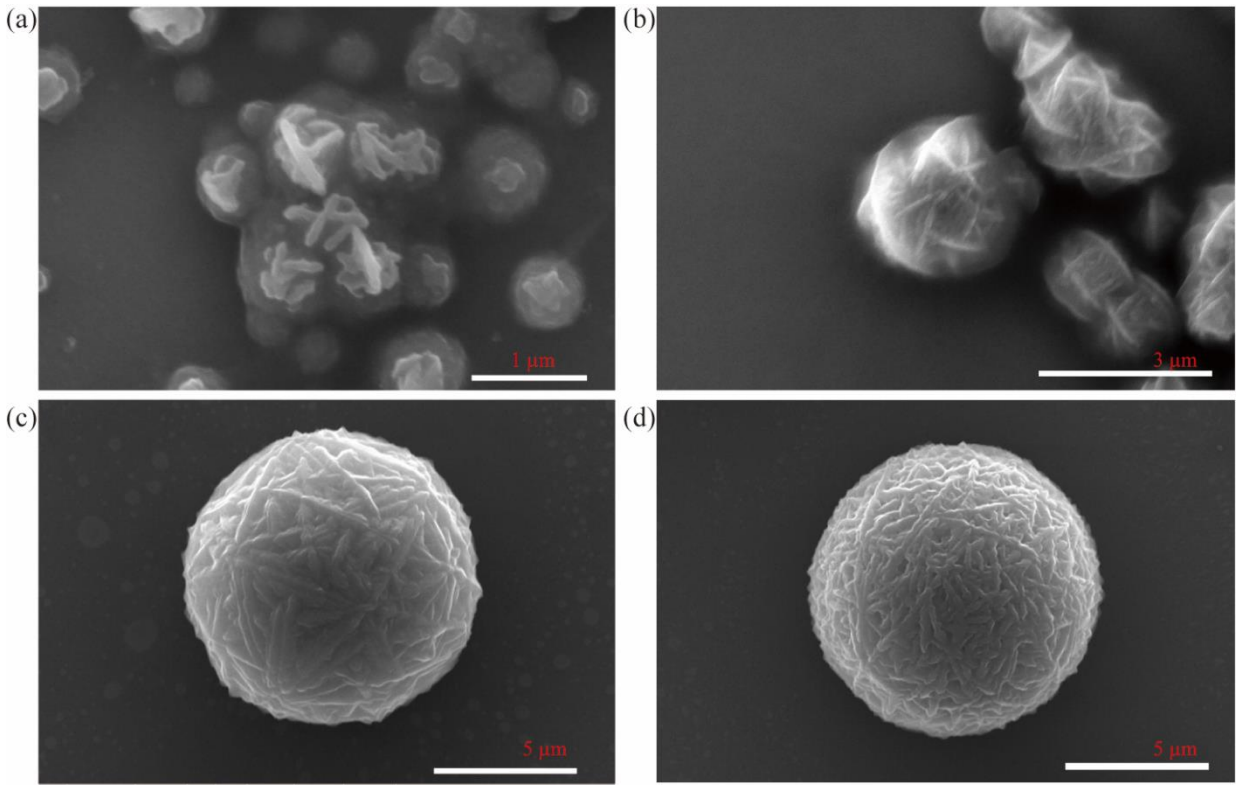

Figure S3. SEM images of polythiophene-C_{60} formed at 313K under different ACN loading ratios (a) 50%, (b) 66.67%, (c) 85.71% and (d) 88.89%.

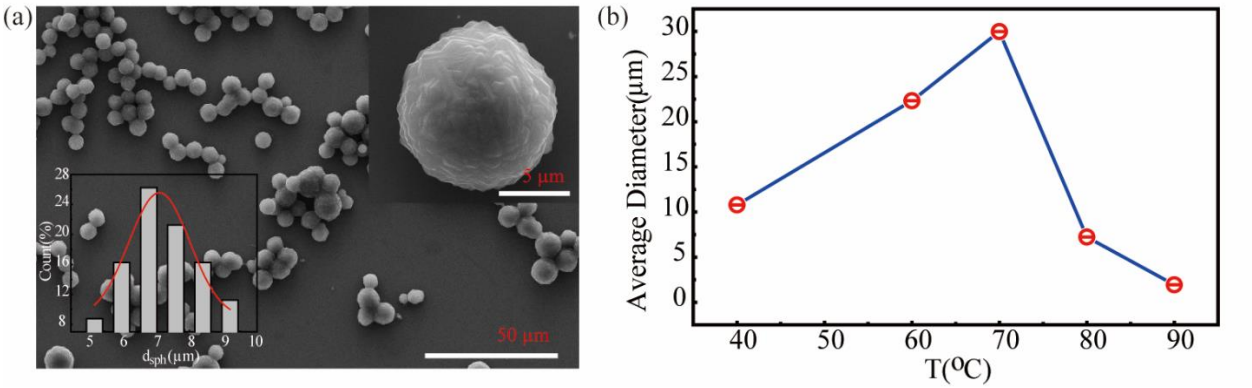

Figure S4. (a) SEM images of three-dimensional polythiophene-C_{60} heterostructure formed under ACN anti-solvent vapor atmosphere at 353K. The inset is high-magnification SEM image and size distribution of the spherical heterostructure. (b) Average size of the spherical crystals under different temperatures.

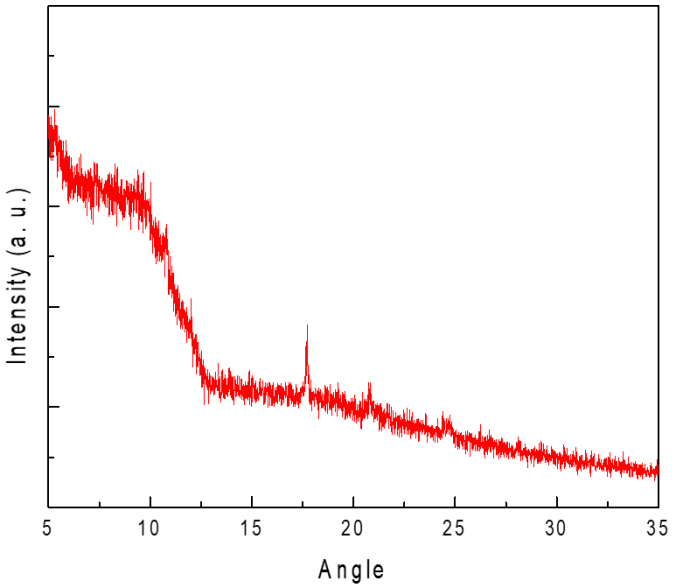

Figure S5. XRD spectrum of polythiophene-C_{60} heterostructures formed at 343K under ACN atmosphere.

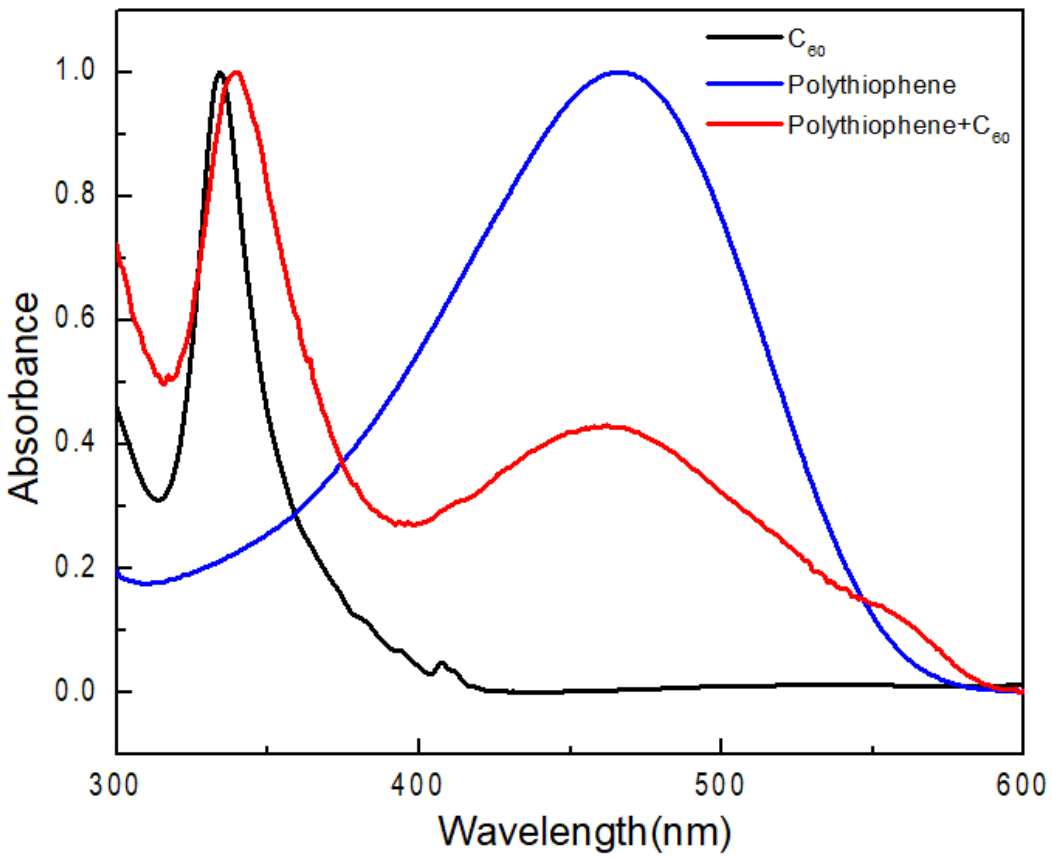

Figure S6. Normalized photoabsorption spectra of C_{60}, polythiophene and polythiophene-C_{60} heterostructures formed at 343K under ACN atmosphere.

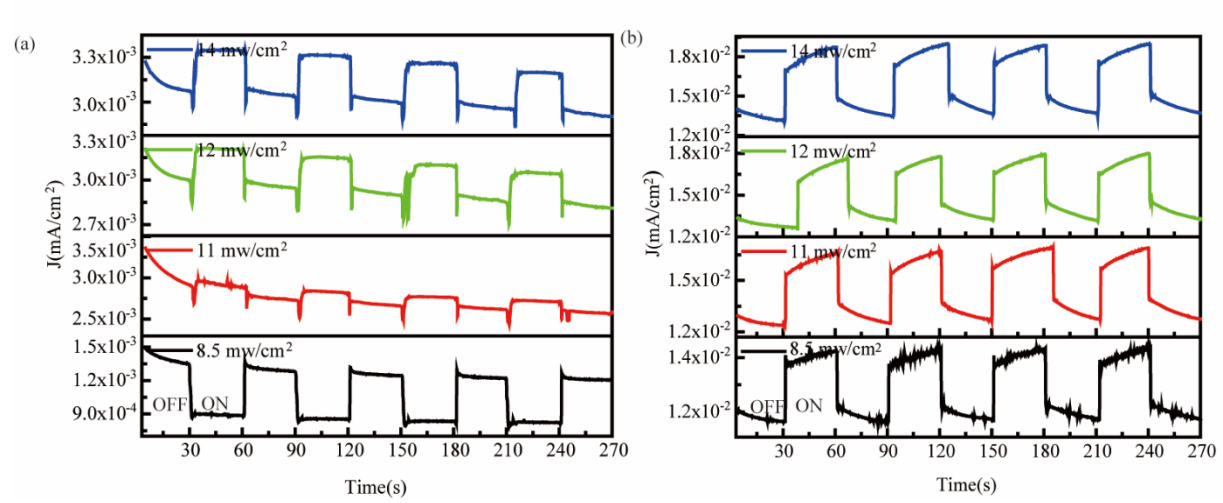

Figure S7. The photoresponse behavior of polythiophene-C_{60} heterostructures formed at (a) 313K and (b) 343K under solar light illumination from 8.5 mw/cm^2 to 14 mw/cm^2 at 2.5 V.

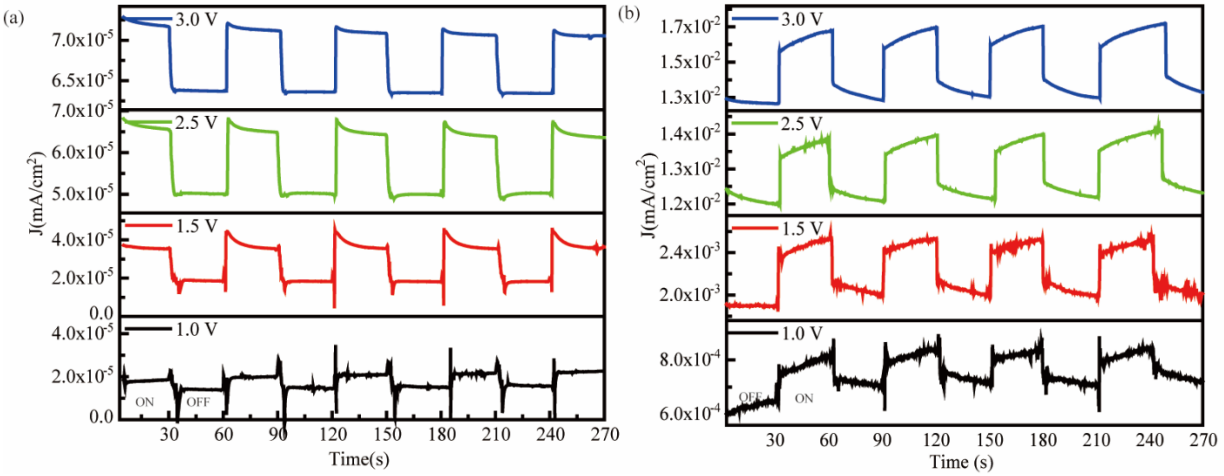

Figure. S8 The photoresponse behavior of polythiophene-C_{60} heterostructures formed at (a) 313K and (b) 343K under external voltage from 1.0 V to 3.0 V and light illumination 12 mw/cm^2.

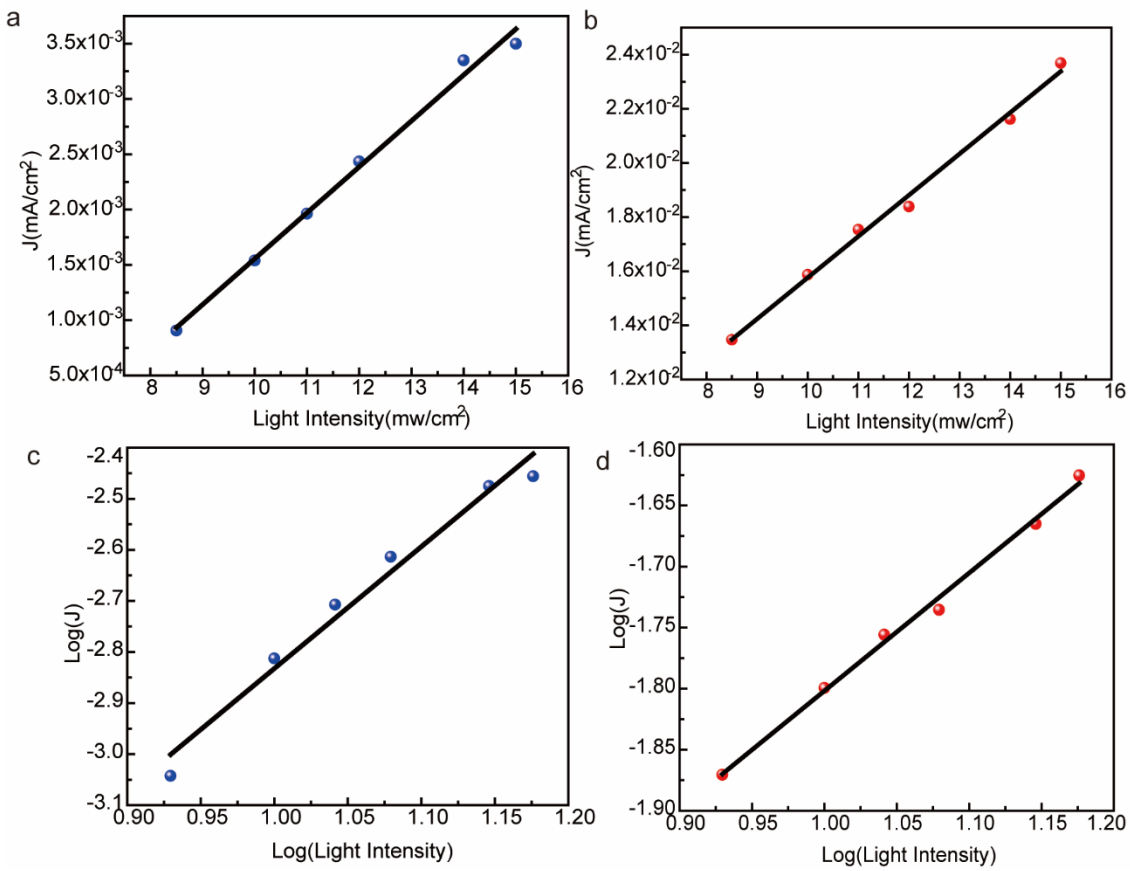

Figure. S9 Photocurrent as a function of light intensity of polythiophene-C_{60} heterostructures formed at (a) 313K and (b) 343K. The corresponding fitting curve using the power law for heterostructures processed at (c) 313K and (d) 343K.

Table 1 Corresponding On/Off ratio and photoresponsibility of polythiophene-C_{60} heterostructures formed at 313K and 343K under different solar light intensities.

Samples	**Polythiophene-C_{60} 313K**		**Polythiophene-C_{60} 343K**	
Light intensity (mw/cm^2)	On-Off ratio (%)	R_i (mA/w)	On-Off ratio (%)	R_i (mA/w)
8.5	22	-0.053	553.96	0.24
10	27.70	0.002	782.35	0.33
11	33.52	0.023	807.81	0.41
12	35.94	0.025	940.94	0.38
14	40.86	0.032	1378.21	0.43
15	44.41	0.033	1555.02	0.15