Supporting Information

Solvent Engineering of Self-Separating Fullerene Crystals for Photodetectors

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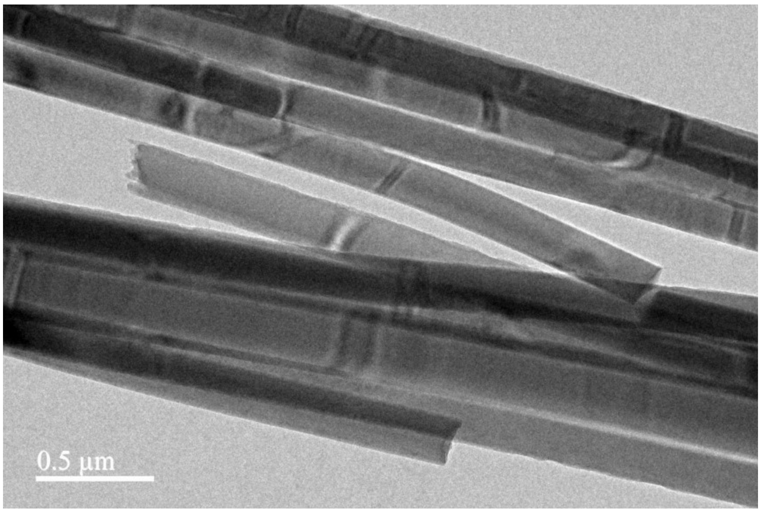


Fig. S1 Additional TEM image of C_{60}NCs_U.

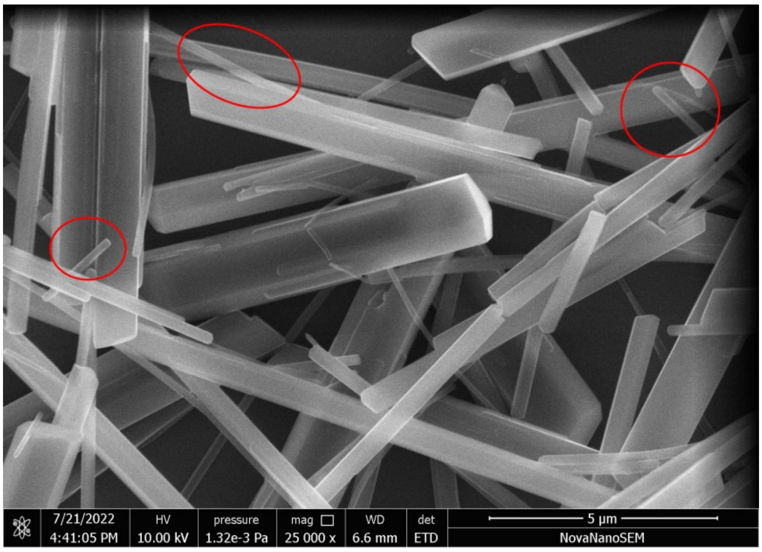


Fig. S2 Additional SEM image of C_{60}NCs_B.

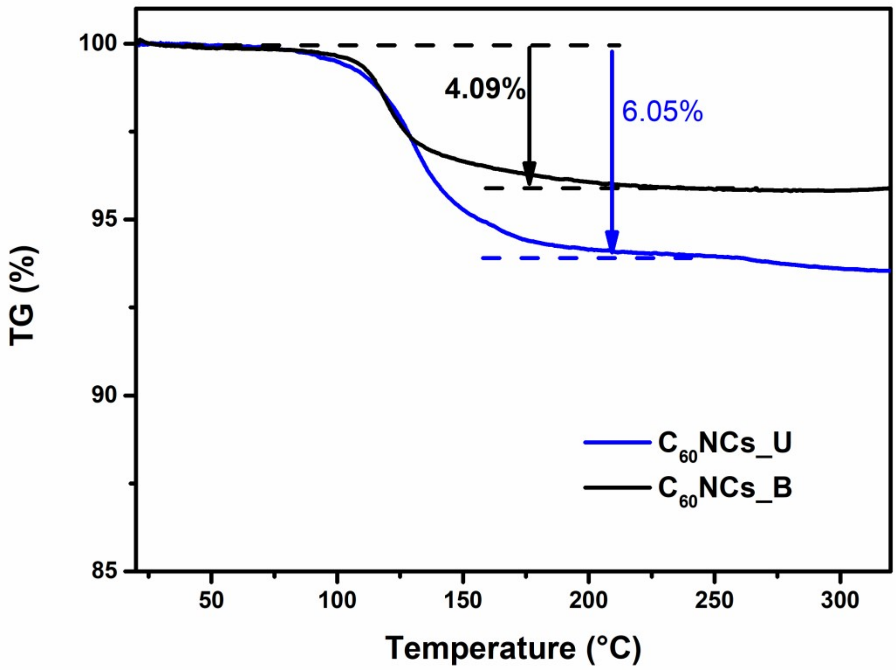
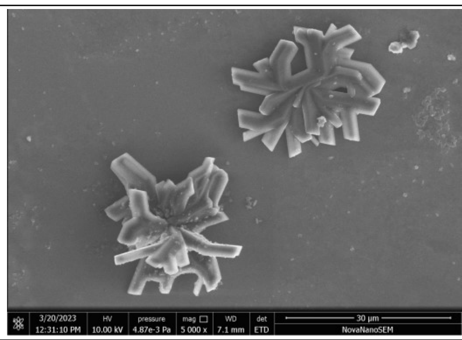
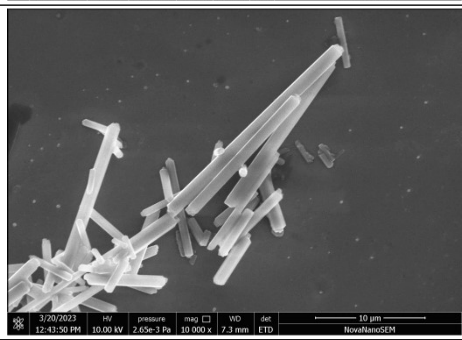
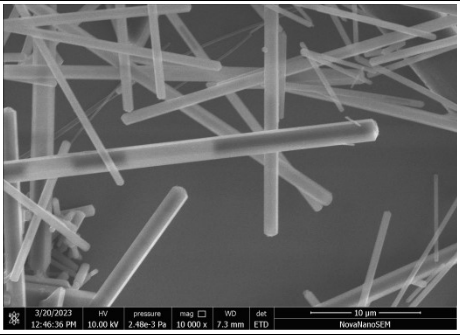
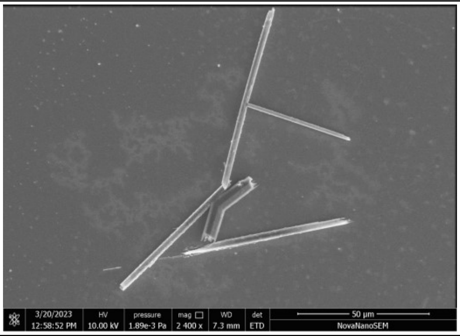


Fig. S3 TG curves of C_{60}NCs_U and C_{60}NCs_B recorded under a nitrogen atmosphere.

Fig. S4 Digital photos of the Self-separating process at different concentration of C_{60} in *m*-Xylene.

Table S1. Digital photos and SEM images of the formation process with different poor solvents.

Sample	Digital photo	SEM image
H_2O		No crystals
DMF		
IPA		
H_2O+DMF		No crystals

H_2O+IPA		
DMF+IPA		

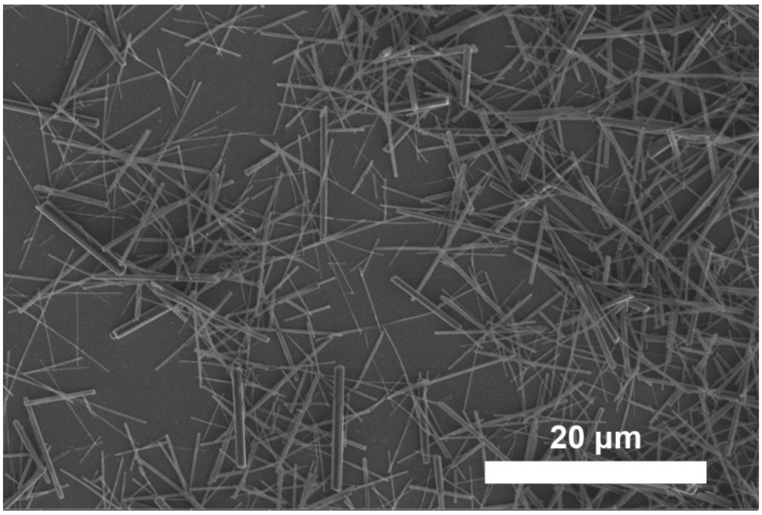


Fig. S5 SEM image of C_{60}NCs_U and C_{60}NCs_B before self-separation process.

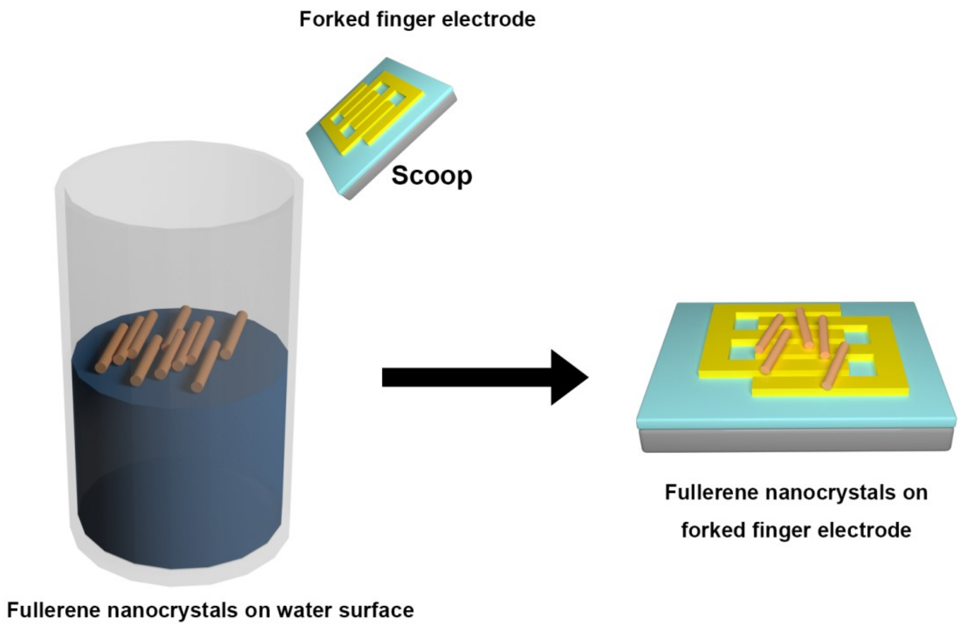


Fig. S6 Scheme illustration of the fullerene nanocrystals transfer on the forked finger electrode from water surface.

Table S2. Statistics of average diameter and average length of C_{60}NCs_U and C_{60}NCs_B before self-separation process.

Sample	Diameter (nm)	Length (μm)
C_{60}NCs_U	205	7.3
C_{60}NCs_B	824	9.5

Table S3. Device currents of C_{60}NCs_U with and without UV illumination at a light density a light density of 0.076, 0.33 and 0.66 mW/cm^2 and at bias of 20, 40 and 60 V respectively.

Light intensity / Voltage	Dark	0.076 mW cm^{-2}	0.33 mW cm^{-2}	0.66 mW cm^{-2}
20	23.6 pA	182 pA	216 pA pA	317 pA
40	27.7 pA	264 pA	405 pA	553 pA
60	65.3 pA	330 pA	627 pA	841 pA

Table S4. Device currents of C_{60}NCs_B with and without UV illumination at a light density a light density of 0.076, 0.33 and 0.66 mW/cm^2 and at bias of 20, 40 and 60 V respectively.

Light intensity / Voltage	Dark	0.076 mW cm^{-2}	0.33 mW cm^{-2}	0.66 mW cm^{-2}
20	58.5 pA	218 pA	262 pA	460 pA
40	101 pA	374 pA	631 pA	951 pA
60	124 pA	517 pA	1190 pA	1890 pA

Table S5. Summary of the main C_{60} nanostructures for photodetectors that have become common in recent years.

Material	Photocurrent	On/off ratio	Ref.
Super-Long fullerene 1D nanostructures	around 80 pA	High	1
Fullerene Microflowers	20 pA	3.2	2
Fullerene Microribbons	103 pA	High	3
Fullerene Single Crystal Microwires (DC-device)	around 150 pA	5.6	4
This work	**Over 1000pA**	**17.08**	

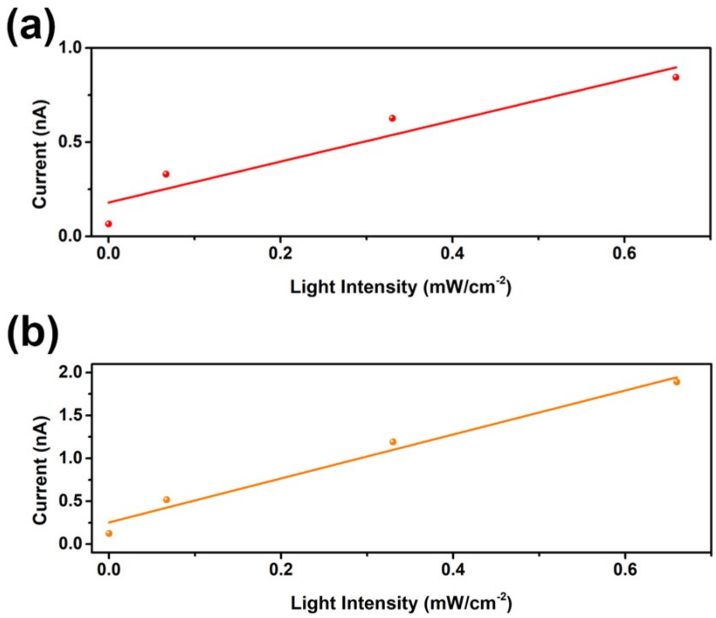


Fig. S7 Under UV irradiation, light-dependence of C_{60}NCs_U and C_{60}NCs_B at 60 V bias.

1. K. Liu, S. Gao, Z. Zheng, X. Deng, S. Mukherjee, S. Wang, H. Xu, J. Wang, J. Liu and T. Zhai, *Adv. Mater.*, 2019, **31**, 1808254.
2. Q. Tang, G. Zhang, B. Jiang, D. Ji, H. Kong, K. Riehemann, Q. Ji and H. Fuchs, *SmartMat*, 2021, **2**, 109-118.
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