

A Quasi-Solid-State Photothermal Supercapacitor via Enhanced Solar Energy Harvest

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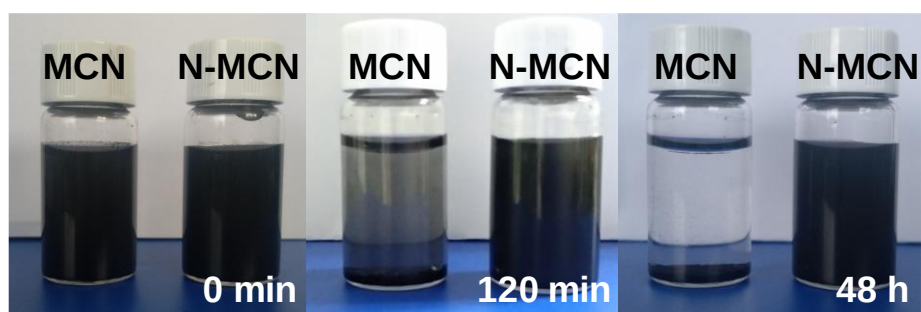


Figure S1. Digital photographs of MCN and N-MCN aqueous solution after standing for different time periods. It showed that N-MCN had enhanced water-dispersity and hydrophilicity/wettability.

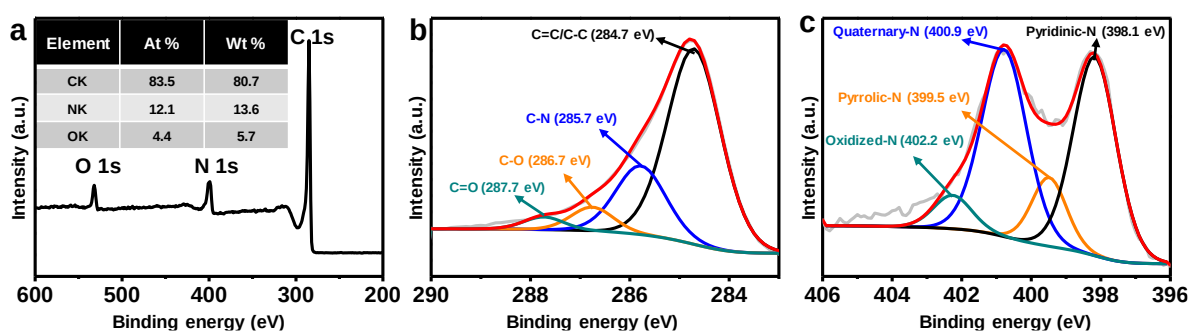


Figure S2. (a) XPS survey spectra, (b, c) C 1s and N 1s spectra of N-MCN. It showed that the N with the dominated Pyridinic-N and Quaternary-N was doped into MCN with plentiful sp²-hybridized carbon framework.

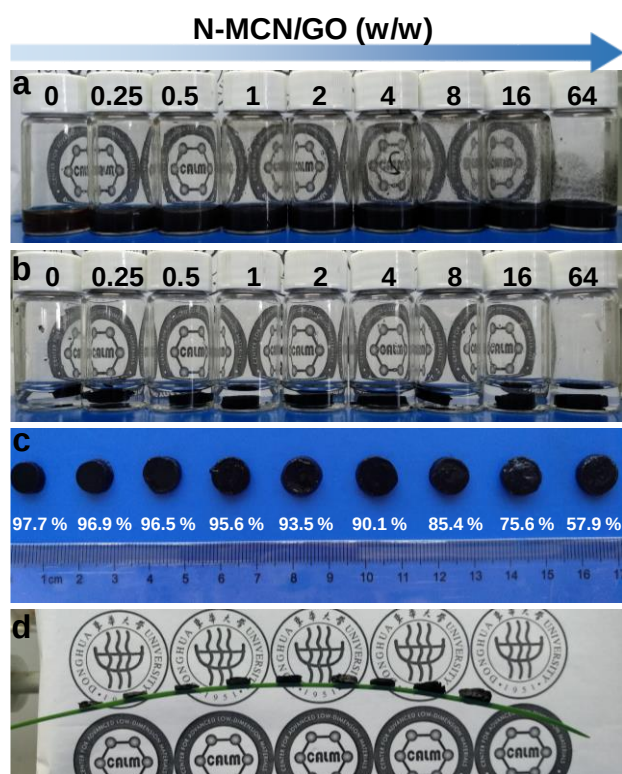


Figure S3. Digital photographs for the facile formation of N-MCN@GH hydrogels and aerogels under a wide N-MCN/GO ratios: (a) N-MCN/GO solution. (b) After hydrothermal treatment. (c) N-MCN@GH hydrogels with various water contents. (c) Light-weight N-MCN@GH aerogels obtained via freeze drying.

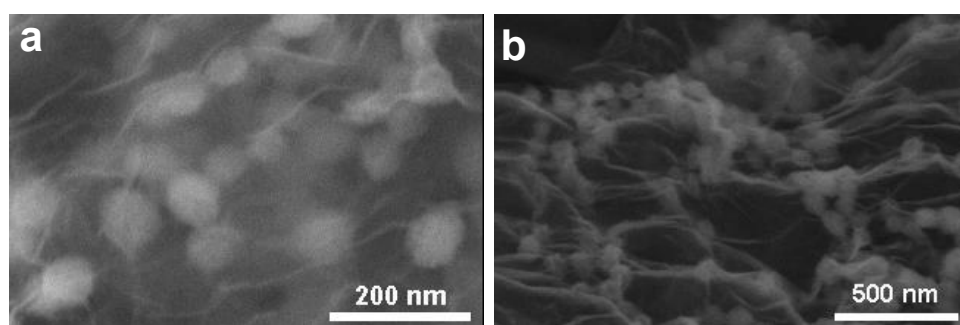


Figure S4. SEM images of N-MCN@GH. It showed that the dispersed N-MCN was homogeneously intercalated between graphene nanosheets to build a 3D porous architecture.

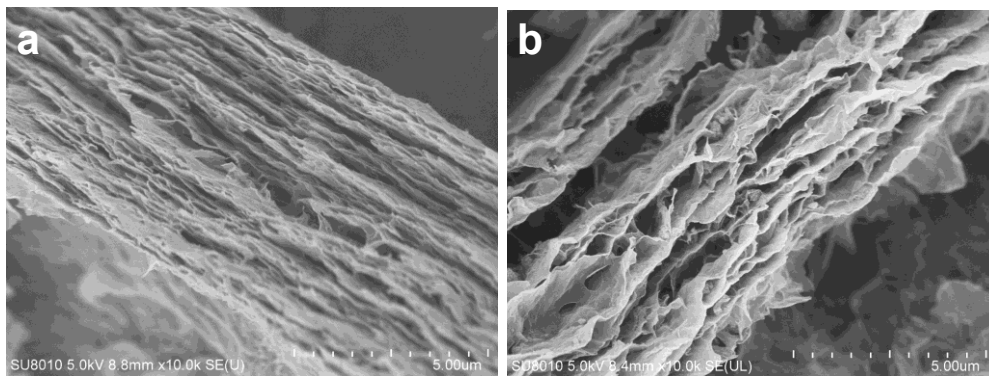


Figure S5. Cross section-view SEM images of (a) GH and (b) N-MCN@GH films. The enlarged interlayer spacing of N-MCN@GH revealed the intercalation of N-MCN between graphene nanosheets.



Figure S6. Photographs of a flexible supercapacitor based on N-MCN@GH film electrode and its compact configuration, with a thickness of 0.25 mm.

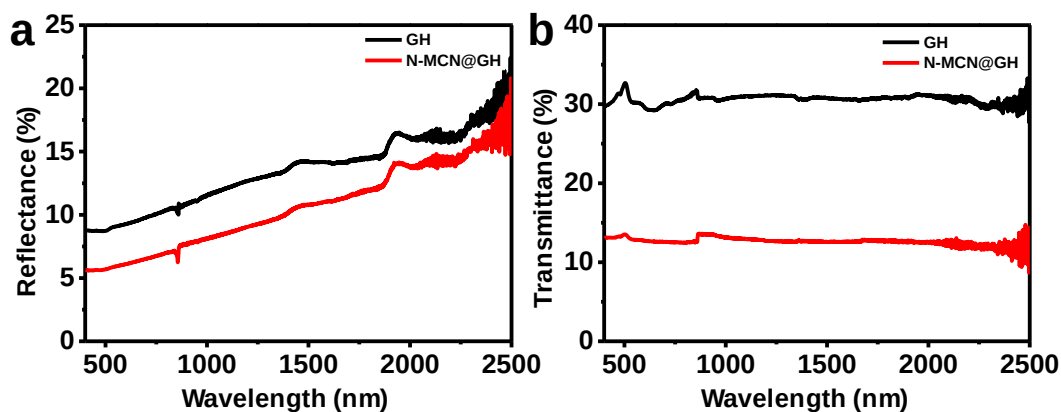


Figure S7. (a) Reflection spectra and (b) Transmission spectra of the GH and N-MCN@GH films. N-MCN@GH had lower reflection and transmission than GH, suggesting the N-MCN intercalation-produced 3D hierarchical porous network for multiple scattering with high absorption.

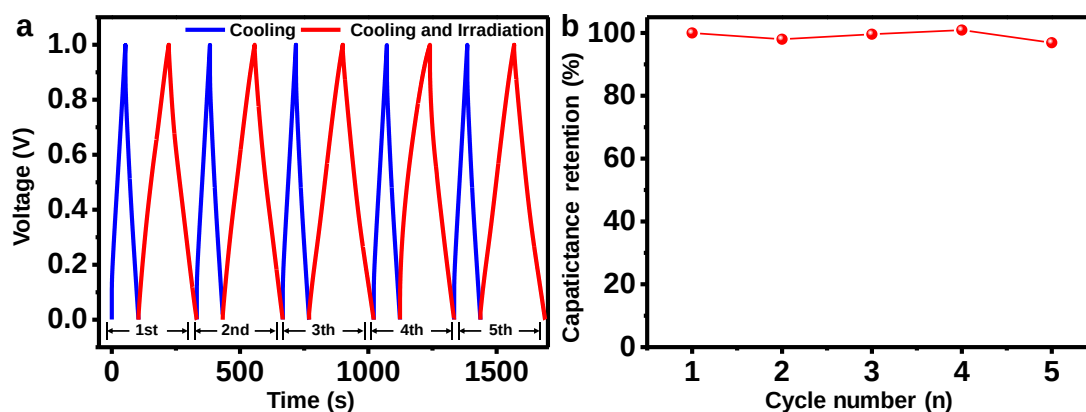


Figure S8. (a) The cycled GCD curves at a current density of 1 A g^{-1} , where blue lines represented the cooling to the 5°C and red lines represented the cooling and irradiation for 10 min. (b) Capacitance retention of N-MCN@GH supercapacitor after different cooling and irradiation cycles as illustrated in “a”.

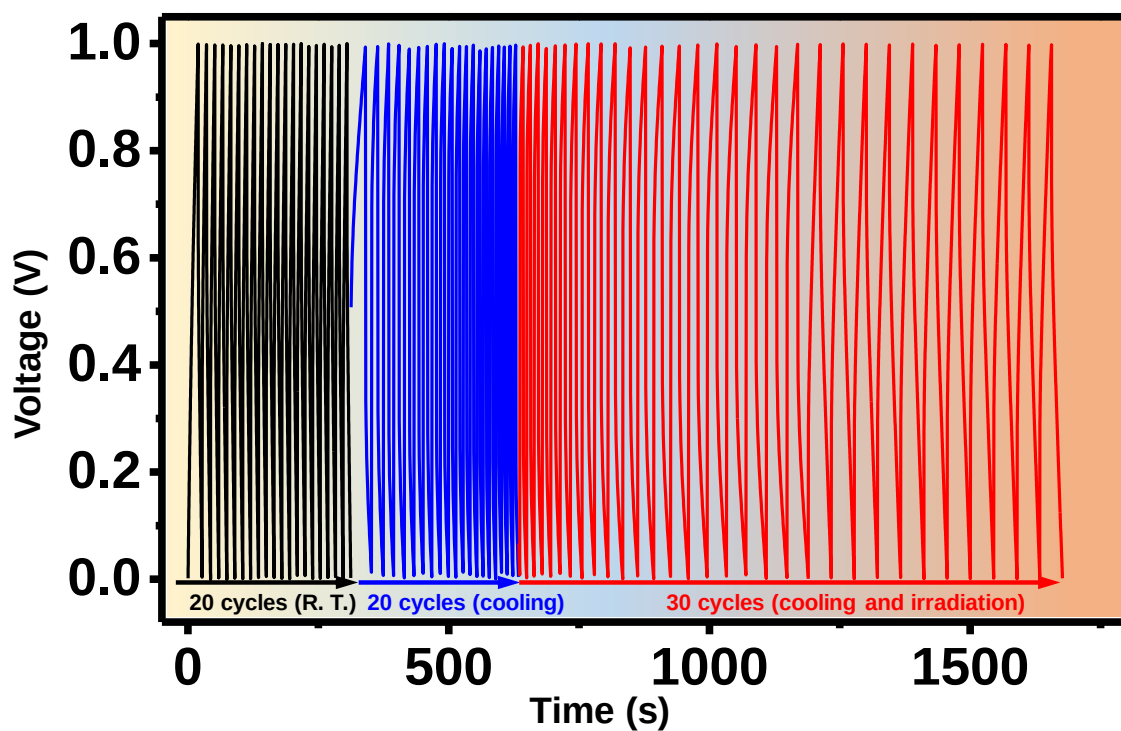


Figure S9. Galvanostatic charge/discharge curves of N-MCN@GH at a current density of 5 A g⁻¹ under different environmental conditions.

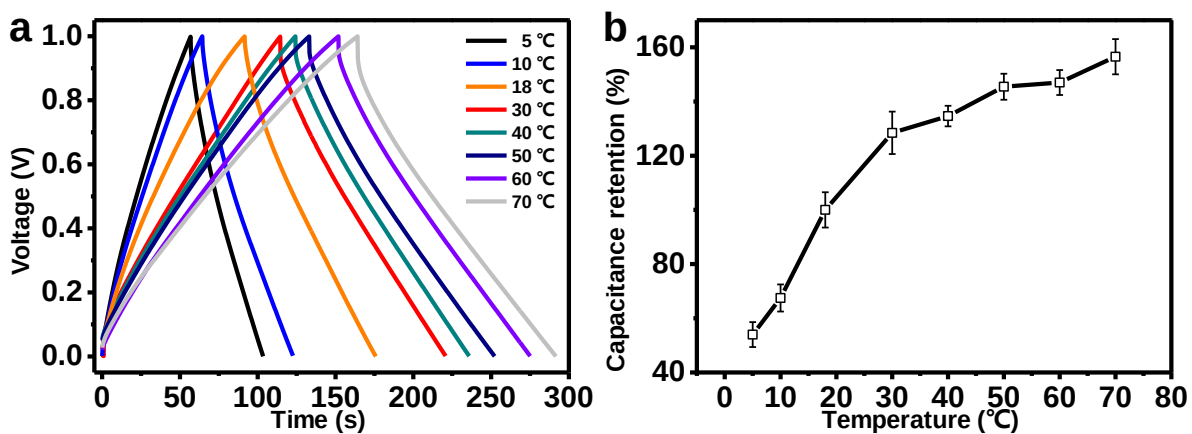


Figure S10. (a) GCD curves at a current density of 1 A g⁻¹ and (b) corresponding capacitance curve of N-MCN@GH supercapacitor at different environment temperatures.

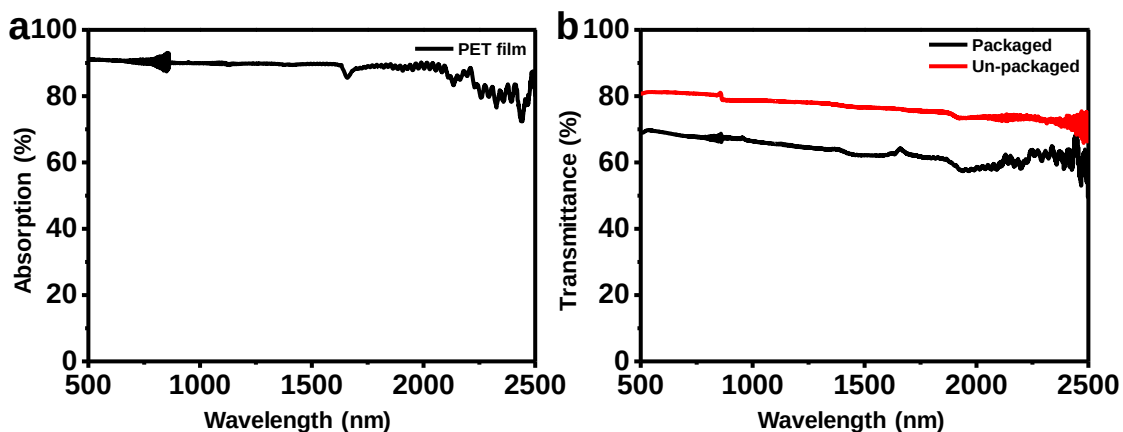


Figure S11. (a) Transmission spectra of the PET films. (b) UV-vis-NIR absorption spectra of N-MCN@GH electrodes with/without PET film packaged.

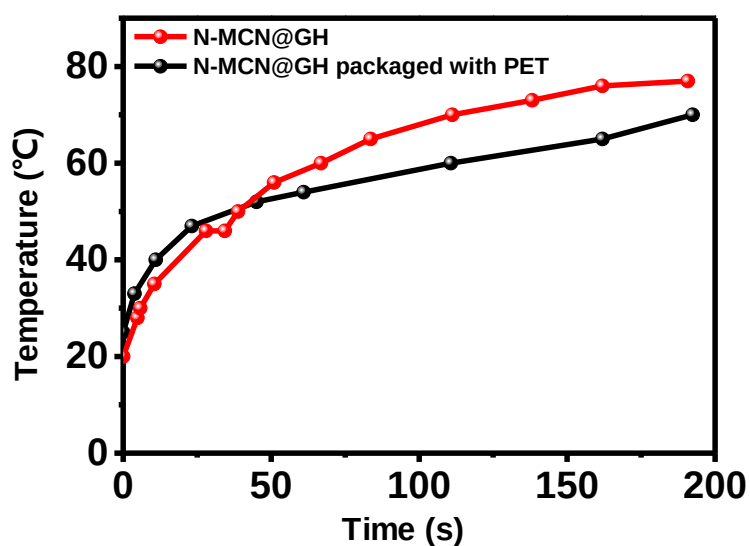


Figure S12. Solar-thermal heating curves N-MCN@GH electrodes with/without PET film packaged under the simulated sunlight illumination (0.36 W/cm^2).

Table S1.
Structural properties of the GH and N-MCN@GH.

Sample	BET surface area ($\text{m}^2 \text{ g}^{-1}$)	Pore volume ($\text{cm}^3 \text{ g}^{-1}$)	Micropore volume ($\text{cm}^3 \text{ g}^{-1}$)	t-Plot Micropore Area ($\text{m}^2 \text{ g}^{-1}$)	Mesopore volume ($\text{cm}^3 \text{ g}^{-1}$)
GH	125	0.53	0.03	62.67	0.52
N-MCN@GH	742	1.19	0.10	187.11	1.11

Table S2.

Comparisons of the electrochemical performance of N-MCN@GH with the reported flexible supercapacitors.

Active material	Electrolyte	Potential window	Volumetric Capacitance	Capacitance retention/ cycle number	Energy density (mWh cm ⁻³)	Power density (mW cm ⁻³)	Ref.
Laser Scribing graphene	PVA/H ₃ PO ₄	1 V	3.67 mF cm ⁻²	96.5%/10000	1.36	200	<i>Science</i> 2012 , 335, 1326-1330
Fe ₂ O ₃ /PPy	PVA/LiCl	1.6 V	382.4 mF cm ⁻² at 0.5 mAcm ⁻²	97%/5000	0.22	165.6	<i>Angew. Chem., Int. Ed.</i> 2017 , 56, 1105.
Ni/MnO ₂	PVA/Na ₂ S O ₄	2.5 V	1.4 F cm ⁻³ at 2.5 mA cm ⁻³	85%/1000	0.78	2.5	<i>ACS Nano</i> 2016 , 10, 1273.
MnO ₂ @NN A//Ppy@NN A	Na ₂ SO ₄	1.6 V	135.15 mF cm ⁻² at 3 A g ⁻¹	106%/20000	1.23	310	<i>Adv. Mater.</i> 2016 , 28, 4105.
rGO/Fe ₂ O ₃	PVA/KOH	1 V	0.25 F cm ⁻³	96%/5000	0.035	0.003	<i>Chemistry of Materials</i> 2017 , 29,14: 6058-65.
Ni-Mn-C	PVA/KOH	0.6 V	0.53 F cm ⁻³	86%/10000	0.027	36.20	<i>J. Mater. Chem. A</i> 2018 , 6,47: 24086-91.
MnO ₂	PVA/LiCl	0.8 V	2 F cm ⁻³ at 0.5A g ⁻¹	91%/20000	0.17	1	<i>Nanoscale</i> 2017 ,9,3593-3600
Graphene	PVA/H ₂ SO ₄	1 V	4.21mF cm ⁻² at 0.41mA cm ⁻²	95%/10000	0.552	561.9	<i>ACS Appl. Mater. Interfaces</i> 2017 , 9, 9763–9771
N- MCN@GH	PVA/H ₂ SO ₄	1 V	8.1F cm ⁻³ 197.5 mF cm ⁻² at 0.64 mA cm ⁻²	80%/10000	1.12	522.08	This work