Supplementary Information

Nacre-like laminate nitrogen-doped porous carbon/carbon nanotubes/graphene composite for excellent comprehensive performance supercapacitors

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Fig. S1. The SEM images of precursors: (a, b) PGMC, (c, d) PGC and (e, f) PMC.



Fig. S2. The TEM images of acid treated MWNTs.



Fig. S3. The TEM images of precursors: (a, b) PGMC, (c, d) PGC and (e, f) PMC.





Fig. S4. The TEM images and nitrogen atom mapping images of (a) PGMC, (b) PGC and (c) PMC.



Fig. S5. The C 1s XPS spectra of PGMC, PGC and PMC.



Fig. S6. Charge/discharge curves of the PGMC, PGC and PMC composite electrodes measured at current density of 1 A g⁻¹ in two-electrode system..



Fig. S7. Charge/discharge curves of composite at different current density: (a) PGMC,(b) PGC and (c) PMC in two-electrode system.



Fig. S8. Charge/discharge curves of composite at different current density: (a) PGMC,

(b) PGC and (c) PMC in three-electrode system.

Sample	N (wt%)	C (wt%)	H (wt%)	O (wt%)
PGMC	17.36	65.52	2.86	14.26
PGC	16.74	68.17	2.85	12.24
РМС	13.88	67.24	2.69	16.19

Table S1. Elemental composition of samples

Table S2. Summary of the specific capacitance of some electrodes

Electrode materials	Gravimetric capacitance (F g ⁻¹)	Volumetric capacitance (F cm ⁻¹)	Scan rate	Electrolyte	Ref.
Graphene nanomesh-carbon nanotube hybrid (GNCN)	294	331	5 mV s ⁻¹	6 M KOH	S1
Binder-free film (HrGO)	251	216	1 A g ⁻¹	6 M KOH	S2
Nitrogen-doped holey graphene (HG)	375	439	0.1 A g ⁻¹	6 M KOH	S3
Compact PANI-CCG film	458	572	5 A g ⁻¹	$1 \text{ M H}_2 \text{SO}_4$	S4

Nitrogen-doped active carbon/graphene (N-AC/Gr)	378.9	257.7	0.05 A g ⁻¹	6 М КОН	S5
Molecular stitching of graphene film (PPD-graphene)	459	711	0.5 A g ⁻¹	1 M H ₂ SO ₄	S6
High-density PANI/graphene composite	546	802	0.5 A g ⁻¹	$1 \text{ M H}_2 \text{SO}_4$	S 7
N,B co-doped graphene	-	488	10 mV s ⁻¹	PVA/H ₂ SO ₄	S 8
Crumpled RGO	396	330	0.40 A cm ⁻ 3	6 M KOH	S9
FGN-300	456	470	0.5 A g ⁻¹	6 M KOH	S10
3D graphene	341	436	1 mV s ⁻¹	6 M KOH	S11
RGO	182	255	1 A g ⁻¹	6 M KOH	S12
EM-CCG	192	256	0.1 A g ⁻¹	$1 \text{ M H}_2 \text{SO}_4$	S13
Porous carbon films	240	220	10 mV s ⁻¹	$0.5 \text{ M} \text{H}_2 \text{SO}_4$	S14
N-doped porous carbon	305	305	2 mV s ⁻¹	6 M KOH	S15
РМС	485	553	1 A g ⁻¹	1 M H ₂ SO ₄	Present work
PGC	538	877	1 A g ⁻¹	1 M H ₂ SO ₄	Present work
PGMC	563	782	1 A g ⁻¹	1 M H ₂ SO ₄	Present work

Table S3. Summary of the electrochemical performance of symmetric supercapacitors

Electrode materials	E _g (W h kg ⁻¹)	P _g (W kg ⁻¹)	E _V (W h L ⁻¹)	P _V (W L ⁻ 1)	Electrolyte	Ref.
High-density PANI/graphene composite	-	-	17.8	29.4	1 M H ₂ SO ₄	S 7
heteroatom-doped porous carbon-tube	-	-	12.15	700	1 M Na ₂ SO ₄	S16
MXene-based fibres	-	-	5.1	1700	PVA/H ₂ S O ₄	S17
N-doped holey graphene/PANI slice	-	-	26.5	175.3	1 M H ₂ SO ₄	S18
graphene composite films by molecular level couplings	-	-	7.18	2.92	PVA/H ₂ S O ₄	S19
Nitrogen-doped active carbon/graphene (N-	13.1	12.5	11.1	10.6	6 М КОН	S5

in aqueous electrolyte.

AC/Gr)						
Solution Processable						
MXene and			3 /	200	PVA/H ₃ P	\$20
Electrochemically	-	-	5.4	200	O_4	320
Exfoliated Graphene						
GHTC-a	8.4	22.5	-	-	6 M KOH	S21
Nitrogen-enriched porous	5.1	10000	_	_	6 М КОН	\$22
carbon nanofiber	5.1	10000	-	-		522
Nitrogen-doped	5 /3	10000	_	_	6 М КОН	\$23
hierarchical porous carbon	5.45	10000	-	-	0 M KOII	525
hollow particle-based						
nitrogen-doped carbon	10.96	250	-	-	$2 \ M \ H_2 SO_4$	S24
nanofibers						
N-YSHMCSs	7.9	1600	-	-	$1 \text{ M H}_2 \text{SO}_4$	S25
PGC	16.4	600	26.7	978	$1 \text{ M H}_2 \text{SO}_4$	Present
100	10.4					work
РМС	13.7	595	15.6	678	1 M H ₂ SO ₄	Present
TWIC		575				work
PGMC	13 72	12997	191	18000	$1 \text{ MH}_2\text{SO}_4$	Present
	13.72		17.1	10000	1 111 112004	work
PGMC	19.8	650	27 51	904	$1 \text{ M H}_2\text{SO}_4$	Present
1 01110	17.0	000	27.01	204	1 111112004	work

Note: E_g : gravimetric energy density, P_g : gravimetric power density, E_v : volumetric energy density, P_v : volumetric power density.

References

- S1 L. Jiang, L. Sheng, C. Long, Z. Fan, Nano Energy 11 (2015) 471–480.
- S2 Y. Bai, X. Yang, Y. He, J. Zhang, L. Kang, H. Xu, F. Shi, Z. Lei, Z.H. Liu, Electrochim. Acta 187 (2016) 543–551.
- S3 Y. Zhang, L. Ji, W. Li, Z. Zhang, L. Lu, L. Zhou, J. Liu, Y. Chen, L. Liu, W. Chen, Y. Zhang, J. Power Sources 334 (2016) 104–111.
- S4 Y. Wang, X. Yang, A.G. Pandolfo, J. Ding, D. Li, Adv. Energy Mater. 6 (2016) 1–6.
- Q. Xie, R. Bao, A. Zheng, Y. Zhang, S. Wu, C. Xie, P. Zhao, ACS Sustain.
 Chem. Eng. 4 (2016) 1422–1430.
- S6 G. Lian, C.C. Tuan, L. Li, S. Jiao, K.S. Moon, Q. Wang, D. Cui, C.P. Wong, Nano Lett. 17 (2017) 1365–1370.
- S7 Y. Xu, Y. Tao, X. Zheng, H. Ma, J. Luo, F. Kang, Q.H. Yang, Adv. Mater.
 27 (2015) 8082–8087.
- S8 Z.S. Wu, K. Parvez, A. Winter, H. Vieker, X. Liu, S. Han, A. Turchanin, X.

Feng, K. Müllen, Adv. Mater. 26 (2014) 4552-4558.

- S9 J.Y. Lee, K.H. Lee, Y.J. Kim, J.S. Ha, S.S. Lee, J.G. Son, Adv. Funct. Mater. 25 (2015) 3606–3614.
- S10 J. Yan, Q. Wang, T. Wei, L. Jiang, M. Zhang, X. Jing, Z. Fan, ACS Nano 8 (2014) 4720–4729.
- S11 J. Hu, Z. Kang, F. Li, X. Huang, Carbon 67 (2014) 221–229.
- S12 Y. Li, D. Zhao, Chem. Commun. 51 (2014) 1–4.
- S13 X. Yang, C. Cheng, Y. Wang, L. Qiu, D. Li, Science. 341 (2013) 534–537.
- S14 Z. Laušević, P.Y. Apel, J.B. Krstić, I. V. Blonskaya, Carbon N. Y. 64 (2013)
 456–463.
- S15 Q. Wang, J. Yan, Z. Fan, Electrochim. Acta 146 (2014) 548–555.
- S16 J. Zhao, Y. Li, G. Wang, T. Wei, Z. Liu, K. Cheng, K. Ye, K. Zhu, D. Cao, Z.
 Fan, J. Mater. Chem. A 5 (2017) 23085–23093.
- S17 S. Seyedin, E. Yanza, J.M. Razal, J. Mater. Chem. A 5 (2017) 24076–24082.
- S18 Z. Fan, Z. Cheng, J. Feng, Z. Xie, Y. Liu, Y. Wang, J. Mater. Chem. A 5 (2017) 16689–16701.
- S19 J. Cao, C. Chen, K. Chen, Q. Lu, Q. Wang, P. Zhou, D. Liu, L. Song, Z. Niu,
 J. Chen, J. Mater. Chem. A 5 (2017) 15008–15016.
- S20 H. Li, Y. Hou, F. Wang, M.R. Lohe, X. Zhuang, L. Niu, X. Feng, Adv. Energy Mater. 7 (2017) 2–7.
- S21 X. Fan, C. Yu, J. Yang, Z. Ling, J. Qiu, Carbon 70 (2014) 130–141.
- K. Huang, M. Li, Z. Chen, Y. Yao, X. Yang, Electrochim. Acta 158 (2015) 305–313.
- S23 G. Wang, J. Zhang, S. Kuang, J. Zhou, W. Xing, S. Zhuo, Electrochim. Acta 153 (2015) 273–279.
- S24 L.F. Chen, Y. Lu, L. Yu, X.W. (David) Lou, Energy Environ. Sci. 10 (2017) 1777–1783.
- S25 C. Liu, J. Wang, J. Li, M. Zeng, R. Luo, J. Shen, X. Sun, W. Han, L. Wang, ACS Appl. Mater. Interfaces 8 (2016) 7194–7204.