Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting Information

Room Temperature Ultrafast Synthesis of N- and O- Rich Graphene Films with Expanded interlayer distance for High Volumetric Capacitance Supercapacitor

Cong Huang[a], Aiping Hu[a]*, Yanhua Li[a], Haifeng Zhou[a], Yali Xu[a], Yan Zhang[a], Shengping Zhou[a], Qunli Tang[a], Chuansheng Chen[b], and Xiaohua Chen[a]*

[a] College of Materials Science and Engineering, Hunan Province Key Laboratory for Advanced Carbon Materials and Applied Technology, Hunan University, Changsha 410082, P. R. China

[b] College of Materials Science and Engineering, Changsha University of Science and Technology, Changsha, 410114, P. R. China

*Corresponding author: xiaohuachen@hnu.edu.cn; hudaaipinghu@126.com

The calculation of the electrochemical measurement results

The gravimetric capacitance was calculated by CV curves as follows:

$$C_g = \frac{\int IdU}{2vm\Delta V} \quad (1)$$

Where C_g (F g^{-1}), v (V s^{-1}), m (g), ΔV (V) and I (A) represents the gravimetric capacitance, the scan rate, the mass of electrode material, the potential window of CV curves, the response-current of CV curves, respectively.

The gravimetric capacitance was calculated by GCD curves using the following equations:

$$C_g = \frac{It}{m\Delta V} \quad (2)$$

(For three electrode system)

$$C_g = \frac{2It}{m\Delta V} \quad (3)$$

(For two electrode system)

Where C_g (F g^{-1}) is the gravimetric capacitance, m (g) is the mass of a single electrode material, ΔV (V) is the potential window of GCD curves (exclude IR drop), I (A) is the discharge current and t is the discharge time (s).

The volumetric capacitance was calculated using the following equation:

$$C_v = C_g \times \rho \quad (4)$$

Where C_v (F cm^{-3}) is the volumetric capacitance, C_g (F g^{-1}) is the gravimetric capacitance and ρ (g cm^{-3}) is the density of electrode.

The volumetric energy density and power density was calculated using the following equation:

$$E_v = \frac{1}{3.6 \times 8} C_V \Delta V^2 \quad (5)$$

$$P_v = \frac{3600E_v}{t} \quad (6)$$

Where E_v (Wh L^{-1}) is the volumetric energy density, C_v (F cm^{-3}) is the volumetric capacitance, ΔV (V) is the potential window (exclude IR drop), P_v (W L^{-1}) is the the volumetric power density and t (s) is the discharge time.

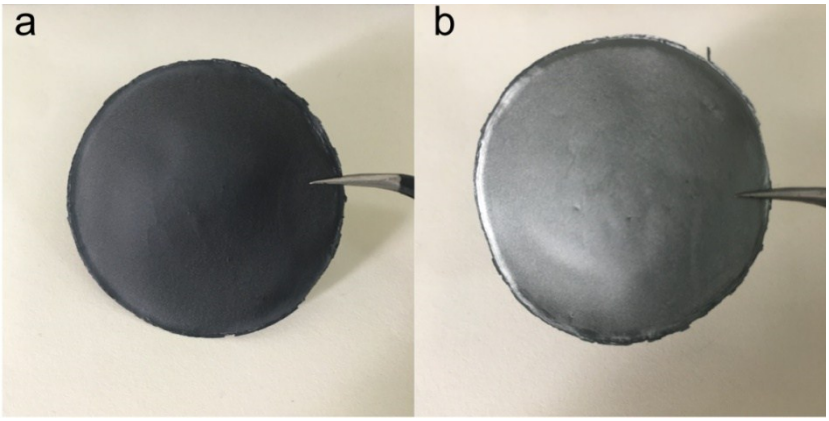

Fig. S1 The digital photo of (a) GO film and (b) RGO film.

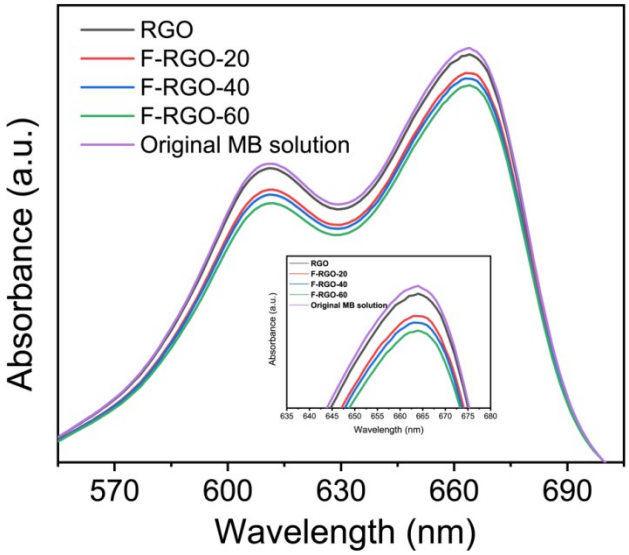

Fig. S2 UV-vis absorption spectra of MB solution after 48 h absorption by RGO and F-RGO-X (X=20, 40, 60).

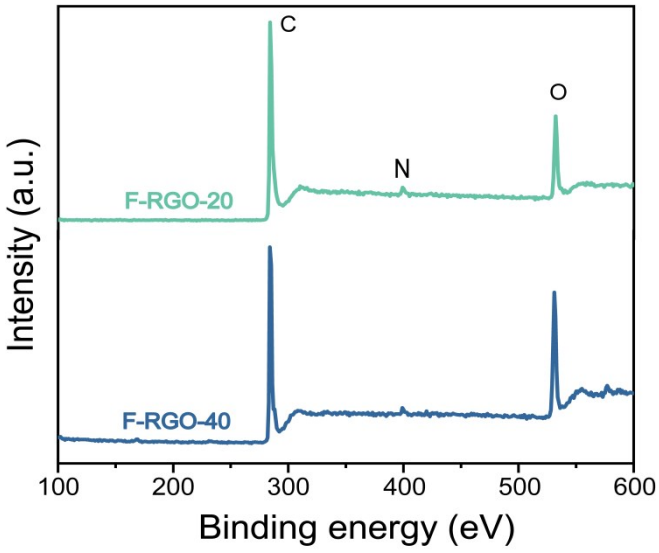

Fig. S3 XPS survey spectra of F-RGO-20 and F-RGO-40.

Table S1. Atomic percentage of different functional groups.

Sample	C=C/C-C	C-O	C=O	O-C=O
RGO	81.47%	13.23%	5.3%	0%
F-RGO-60	69.27%	21.67%	7.04%	2.02%

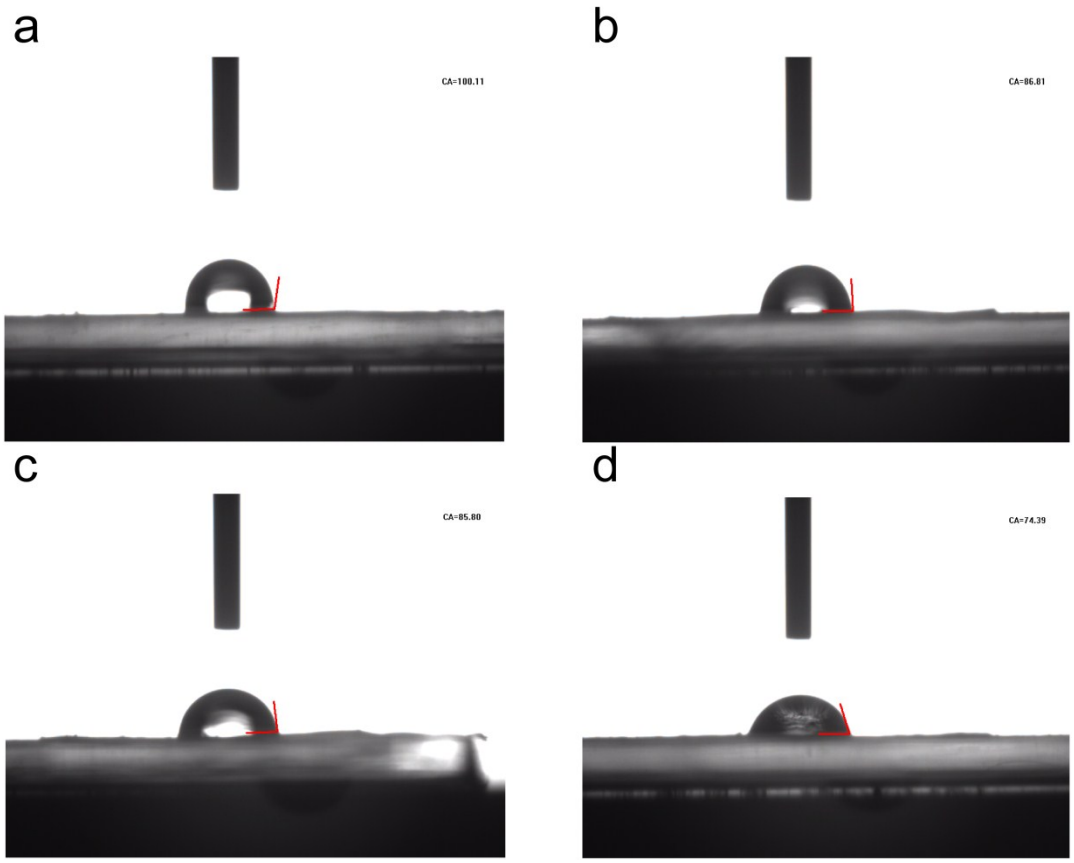

Fig. S4 Water contact angle of (a) RGO, (b) F-RGO-20, (c) F-RGO-40, (d) F-RGO-60.

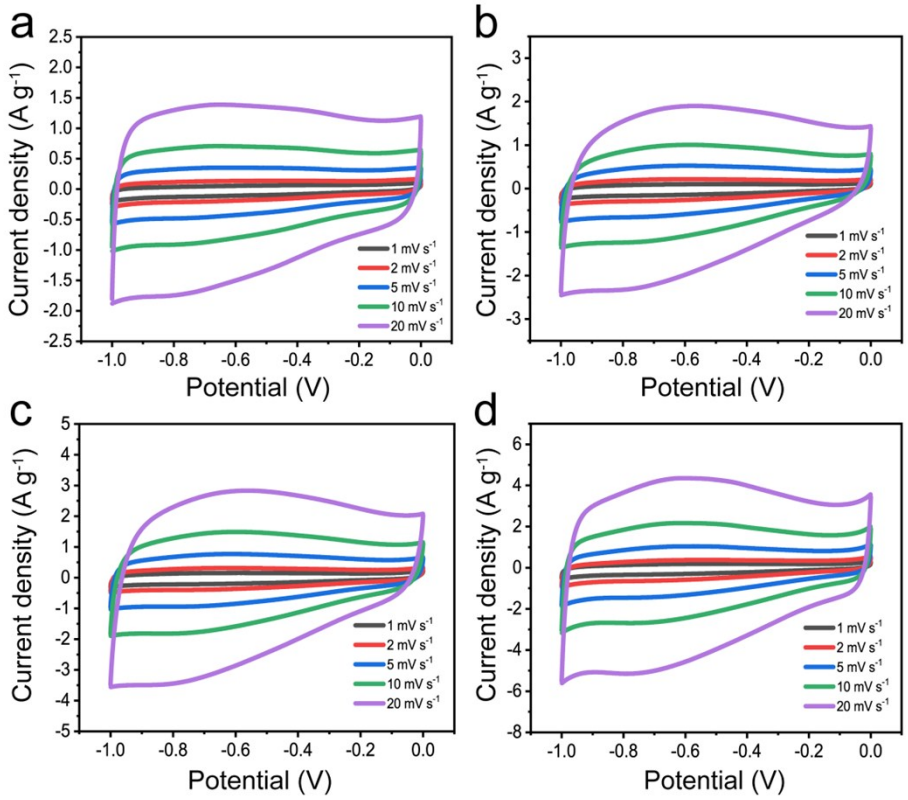

Fig. S5 CV curves of (a) RGO, (b) F-RGO-20, (c) F-RGO-40, (d) F-RGO-60 at different scan rate.

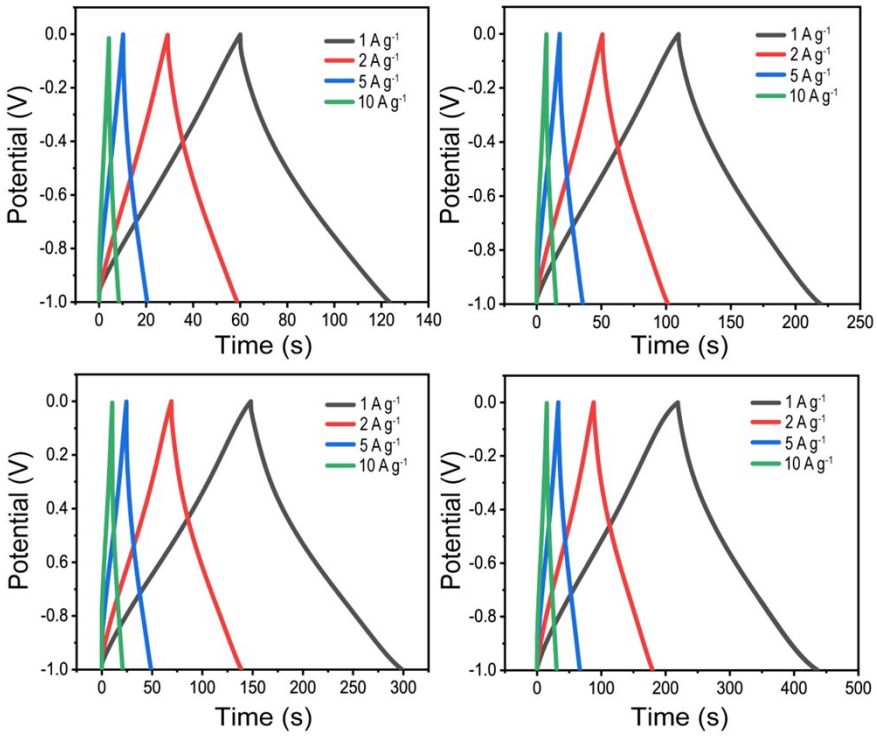

Fig. S6. GCD curves of (a) RGO, (b) F-RGO-20, (c) F-RGO-40, (d) F-RGO-60 at different current density.

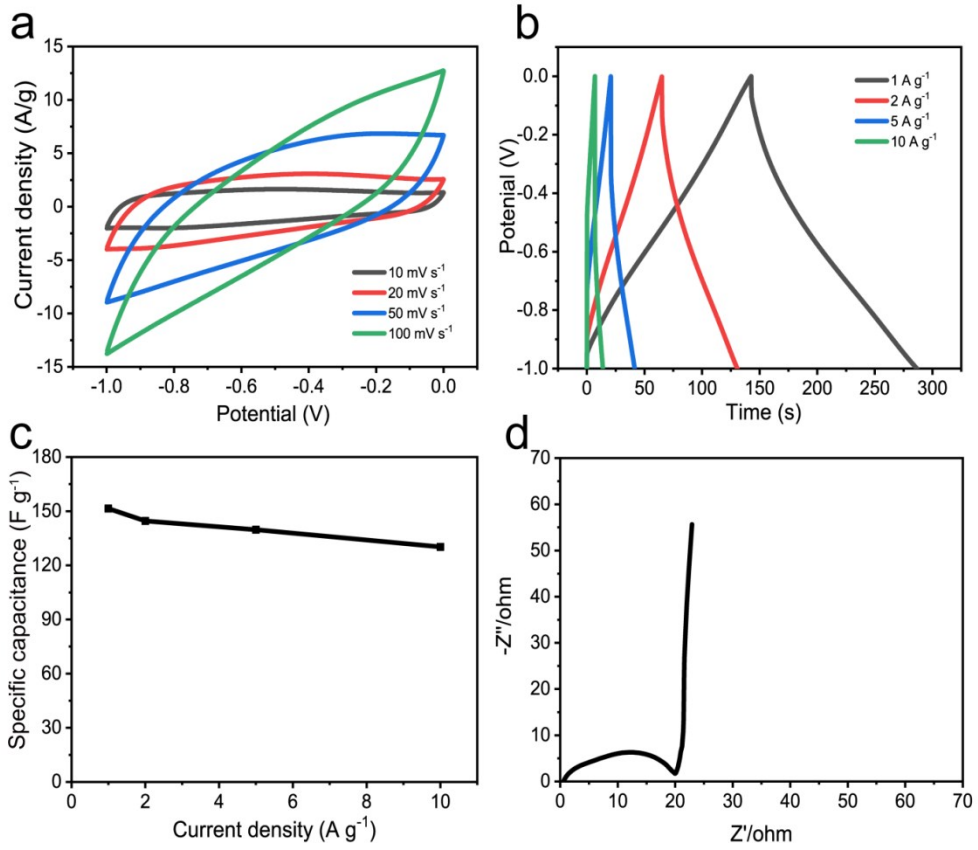

Fig. S7 (a) CV curves of F-RGO-80 at different scan rates. (b) GCD curves of F-RGO-80 at different current densities. (c) Comparison of specific capacitances versus current densities. (d) Nyquist plots of F-RGO-80.

Table S2. Comparison of electrochemical performances of graphene based materials in aqueous electrolyte.

Materials	Density ($g\ cm^{-3}$)	Electrolyte	Test	C_g ($F\ g^{-1}$)	C_v ($F\ cm^{-3}$)
Ternary-doped holey graphene hydrogel[1]	0.67	1.0 M H_2SO_4	1 A g^{-1}	350	234★
Graphene–carbon nanosphere films[2]	1.4	6.0 M KOH	1 A g^{-1}		252
Graphene–CNT films[3]	1.5	6.0 M KOH	1 A g^{-1}		250
the reduced holey graphene films[4]	1.14	1.0 M H_2SO_4	1 A g^{-1}	260	297★
Activated carbon-graphene[5]	0.76	6.0 M KOH	1 A g^{-1}		120
Folded Graphene Ribbon Film[6]	0.92	6.0 M KOH	2 mV s^{-1}	318	293★
Iodine-steam doped graphene films[7]		6.0 M KOH	0.2 A g^{-1}	150	
Carbon fiber-graphene[8]	0.00075	1.0 M H_2SO_4	1 A g^{-1}	215	0.161★
Defect-enriched graphene block[9]	0.917	6.0 M KOH	1 A g^{-1}	235	215★
3D porous RGO film[10]	0.95	1.0 M H_2SO_4	1 A g^{-1}	181.3	172.3
This work	**1.47**	**6.0 M KOH**	**1 A g^{-1}**	**217.3**	**319.4★**
			0.1 A g^{-1}	**178.6**	**262.5**

★represent the three-electrode system

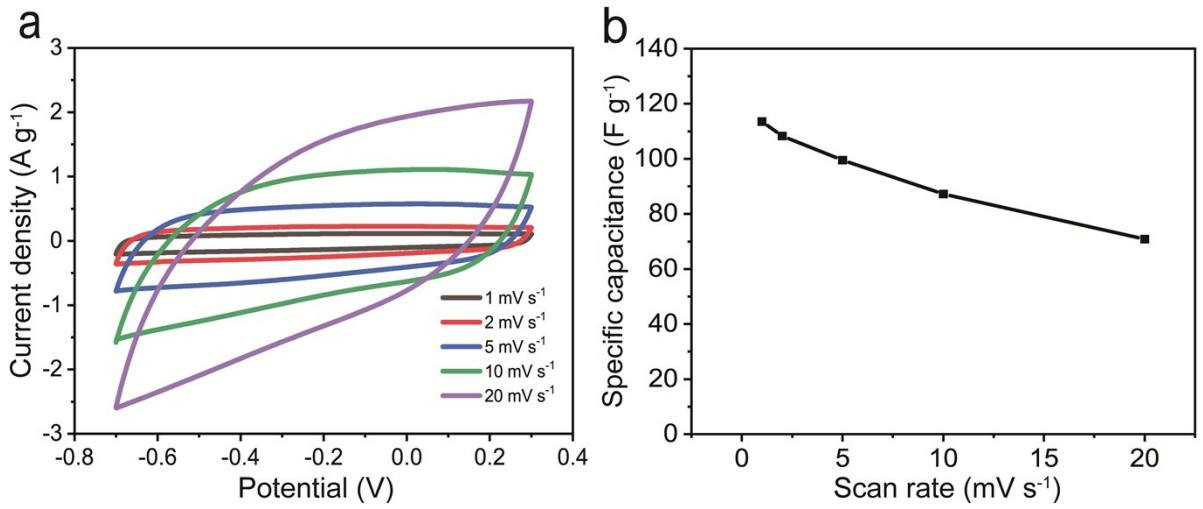

Fig. S8 (a) CV curves of F-RGO-60 in 0.5 M Na_2SO_4 with a three-electrode configuration. (b) Comparison of specific capacitances versus scan rates.

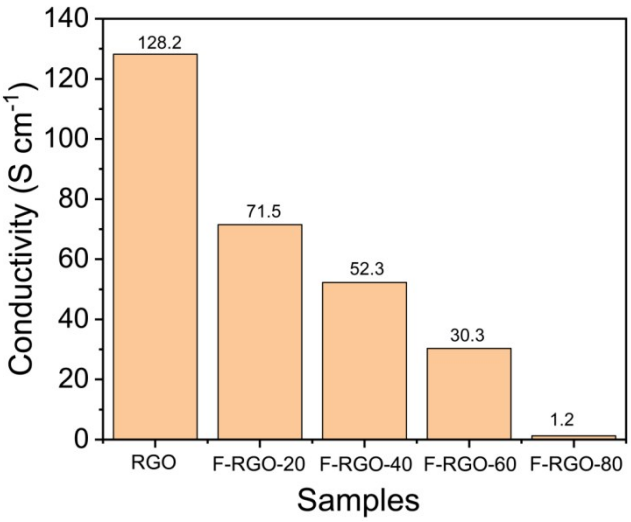

Fig. S9 Electrical conductivity of RGO film and F-RGO-X (X=20, 40, 60 and 80).

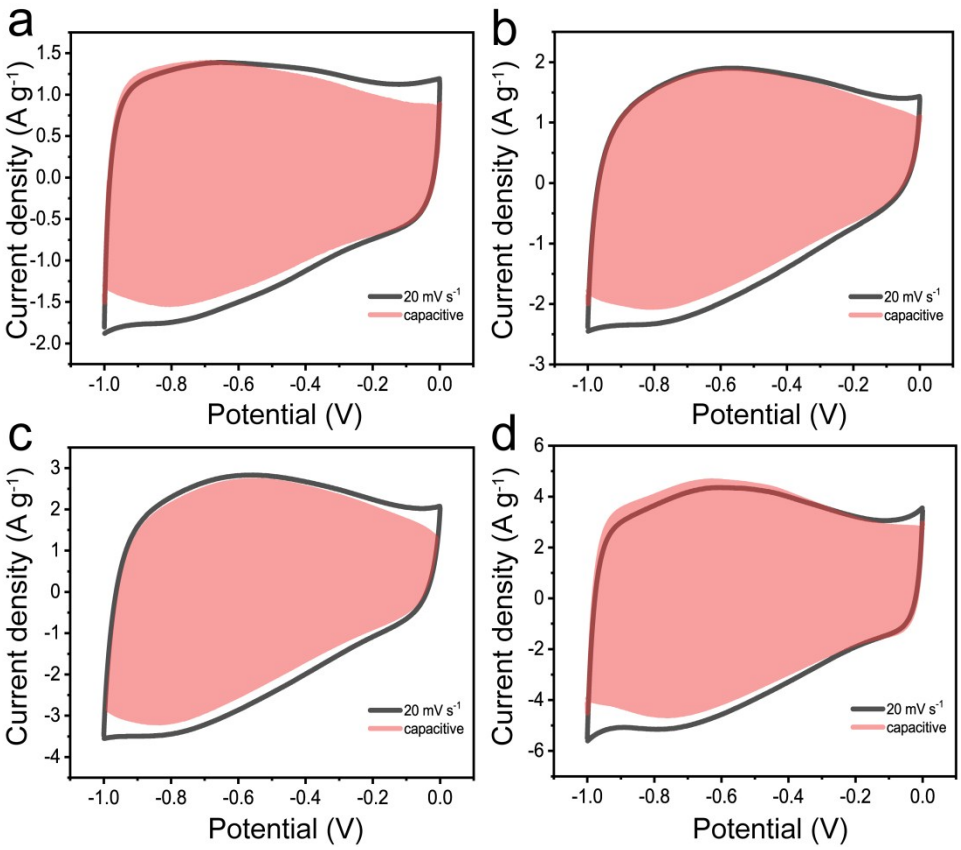

Fig. S10 Capacitive contribution of (a) RGO, (b) F-RGO-20, (c) F-RGO-40, (d) F-RGO-60 at the scan rate of 20 mV s^{-1}.

References

1. Z. H. Pan, H. Z. Zhi, Y. C. Qiu, J. Yang, L. D. Xing, Q. C. Zhang, X. Y. Ding, X. S. Wang, G. G. Xu, H. Yuan, M. Chen, W. F. Li, Y. G. Yao, N. Motta, M. N. Liu and Y. G. Zhang, *Nano Energy*, 2018, **46**, 266-276.
2. N. Díez, M. Qiao, J. L. Gómez-Urbano, C. Botas, D. Carriazo and M. M. Titirici, *Journal of Materials Chemistry A*, 2019, **7**, 6126-6133.
3. N. Díez, C. Botas, R. Mysyk, E. Goikolea, T. Rojo and D. Carriazo, *Journal of Materials Chemistry A*, 2018, **6**, 3667-3673.
4. D. Q. Liu, Q. W. Li and H. Z. Zhao, *Journal of Materials Chemistry A*, 2018, **6**, 11471-11478.
5. P. Li, H. Li, D. Han, T. Shang, Y. Deng, Y. Tao, W. Lv and Q. H. Yang, *Advanced Science*, 2019, DOI: 10.1002/advs.201802355.
6. L. Z. Sheng, J. Chang, L. L. Jiang, Z. M. Jiang, Z. Liu, T. Wei and Z. J. Fan, *Advanced Functional Materials*, 2018, **28**.
7. Y. C. Zhu, X. K. Ye, H. D. Jiang, L. L. Wang, P. Zhao, Z. Y. Yue, Z. Q. Wan and C. Y. Jia, *Journal of Power Sources*, 2018, **400**, 605-612.
8. H. L. Luo, P. X. Xiong, J. Xie, Z. W. Yang, Y. Huang, J. M. Hu, Y. Z. Wan and Y. H. Xu, *Advanced Functional Materials*, 2018, **28**.
9. Y. Dong, S. Zhang, X. Du, S. Hong, S. Zhao, Y. Chen, X. Chen and H. Song, *Advanced Functional Materials*, 2019, DOI: 10.1002/adfm.201901127.
10. Y. Shao, M. F. El-Kady, C. W. Lin, G. Zhu, K. L. Marsh, J. Y. Hwang, Q. Zhang, Y. Li, H. Wang and R. B. Kaner, *Adv Mater*, 2016, **28**, 6719-6726.