

Supporting information

Regulating oxygen functionalities of cellulose-derived hard carbon toward superior sodium storage

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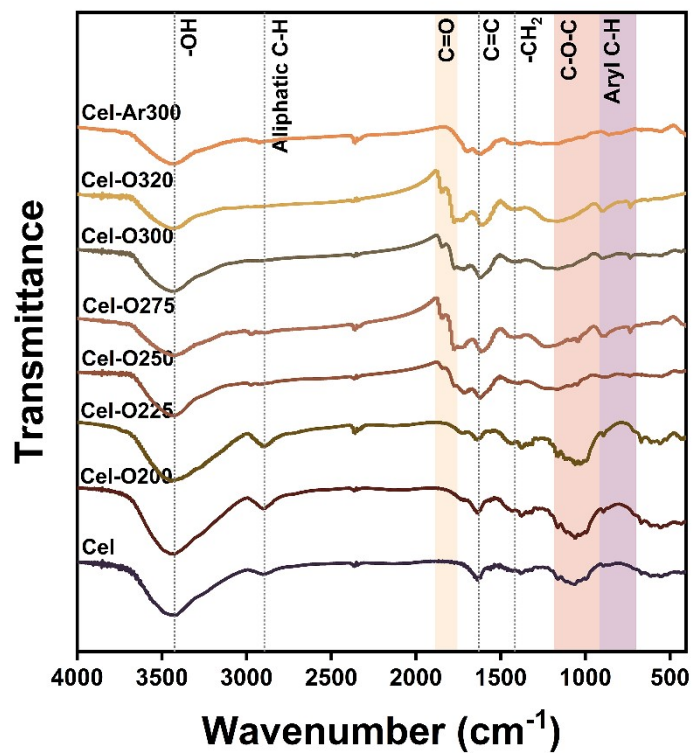


Fig. S1. FTIR spectra of Cel-Ox.

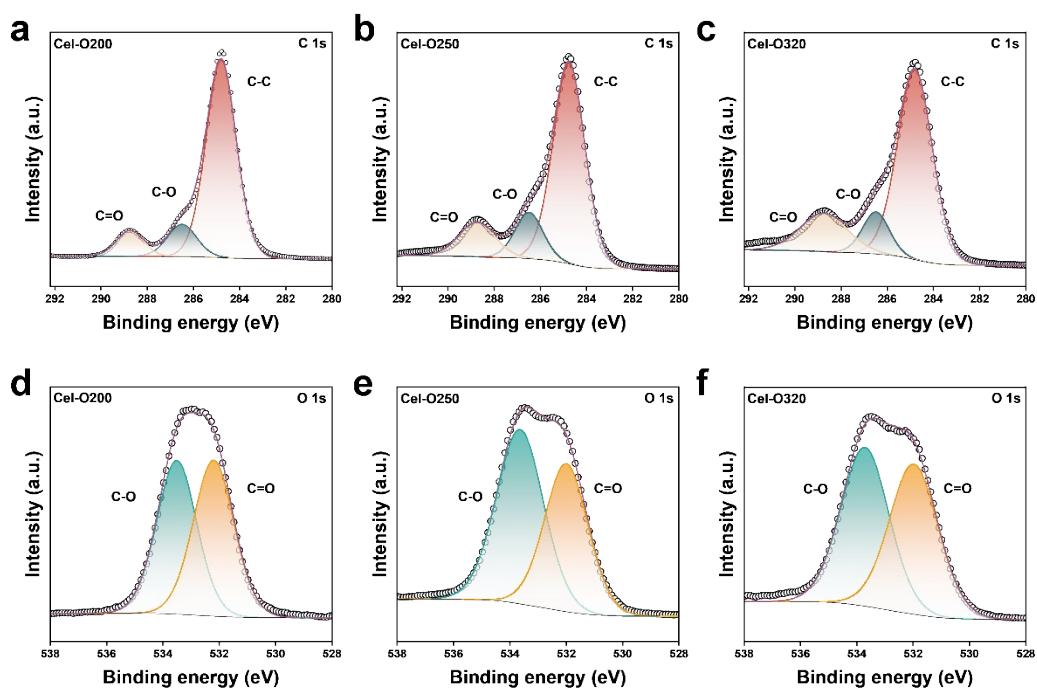


Fig. S2. High resolution C 1s and O 1s spectra of Cel-O200, Cel-O250, and Cel-O320.

Table S1. The content of the C 1s and O 1s peaks in Cel-Ox.

B.E. (eV)	C1 (284.80)	C2 (286.49)	C3 (284.75)	O1 (531.98)	O2 (533.51)
Assignment	C–C	C–O	C=O	C=O	C–O
Cel-O200	78.41	12.41	9.81	49.22	50.78
Cel-O250	71.55	14.53	13.92	56.11	43.89
Cel-O300	77.13	13.49	9.83	57.13	42.87
Cel-O320	69.45	11.75	18.80	51.13	48.87
Cel-Ar300	79.07	14.43	6.50	46.10	53.90

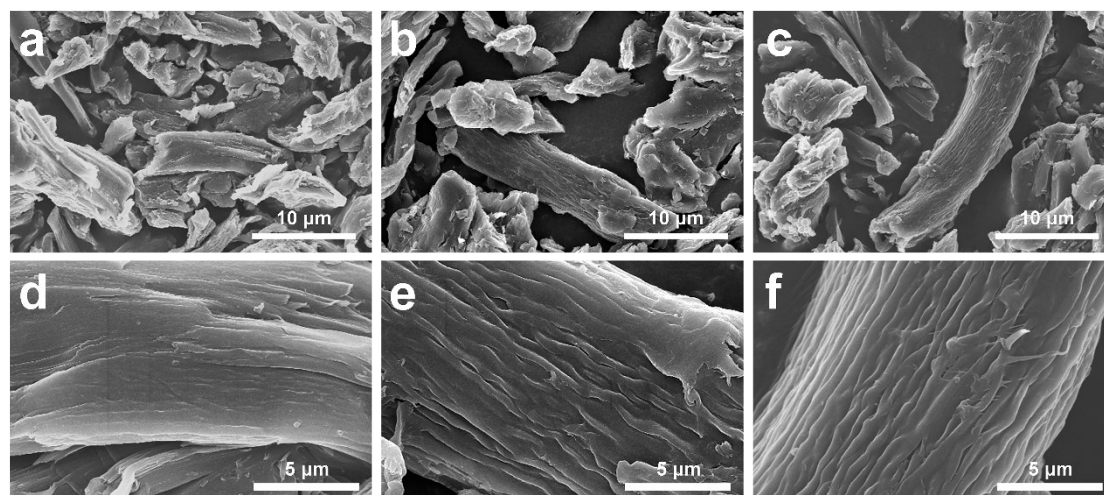


Fig. S3. SEM images of (a, d) Cel-O200-1500, (b, e) Cel-O250-1500, (c, f) Cel-O320-1500.

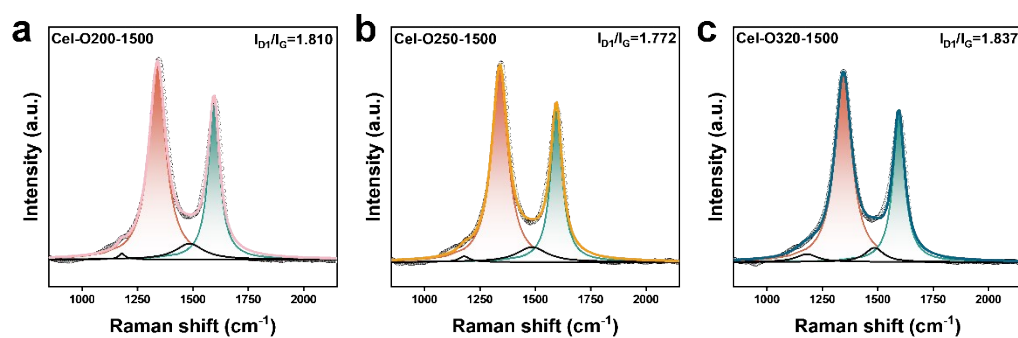


Fig. S4. Raman spectra of Cel-O200-1500, Cel-O250-1500, Cel-O320-1500.

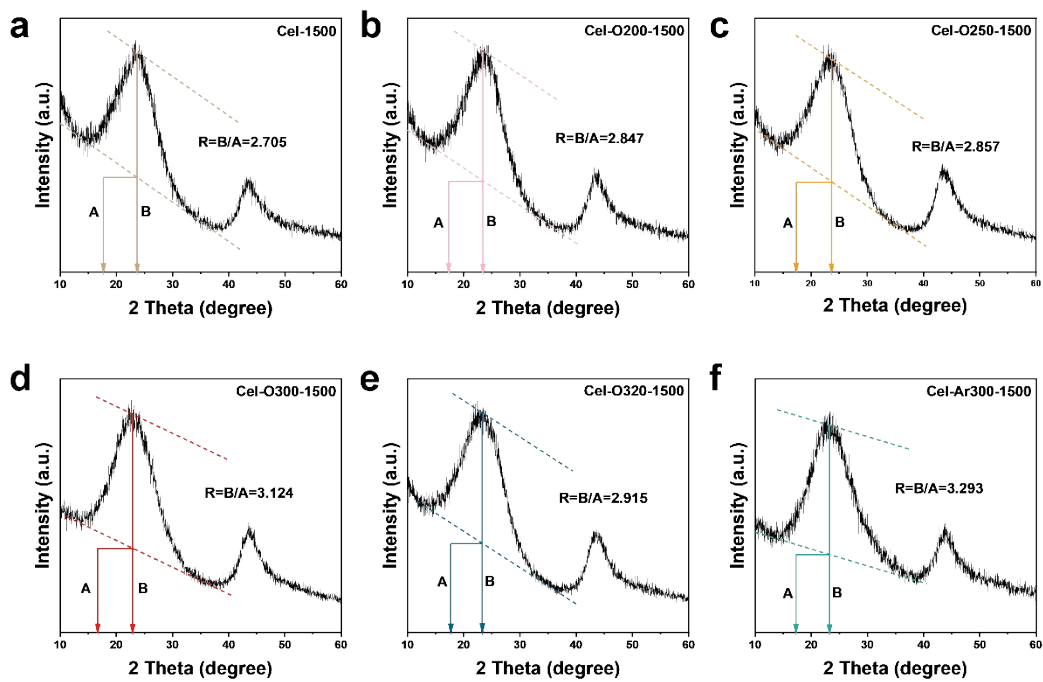


Fig. S5. Calculation of R values of Cel-1500, Cel-Ox-1500, and Cel-Ar300-1500.

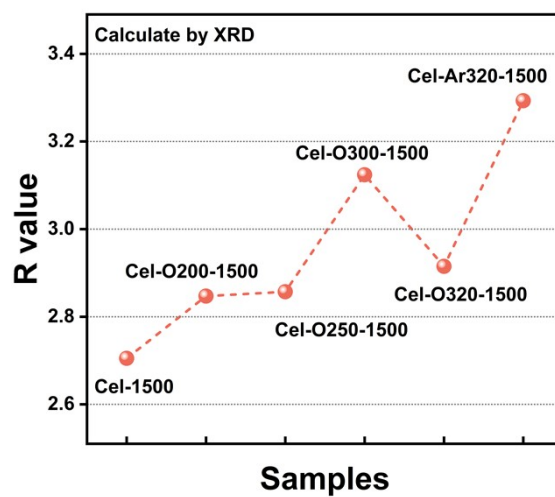


Fig. S6. The variation pattern of R values.

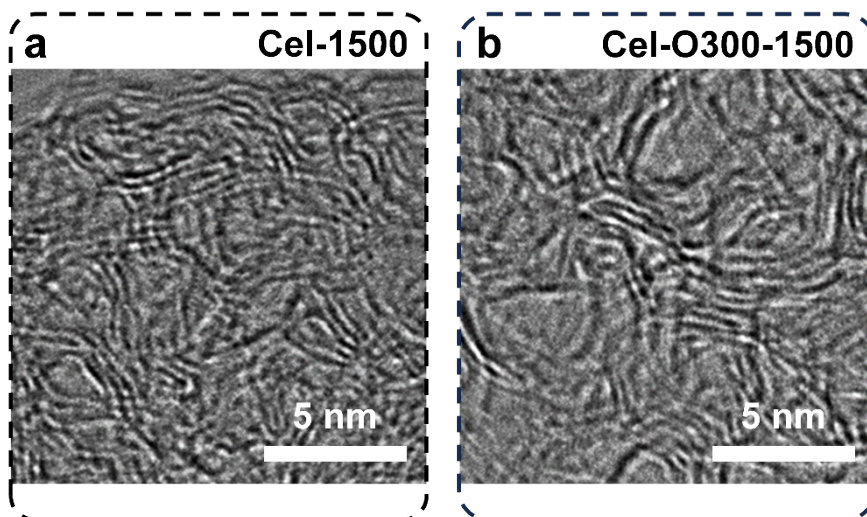


Fig. S7. HRTEM images of Cel-1500 and Cel-O300-1500.

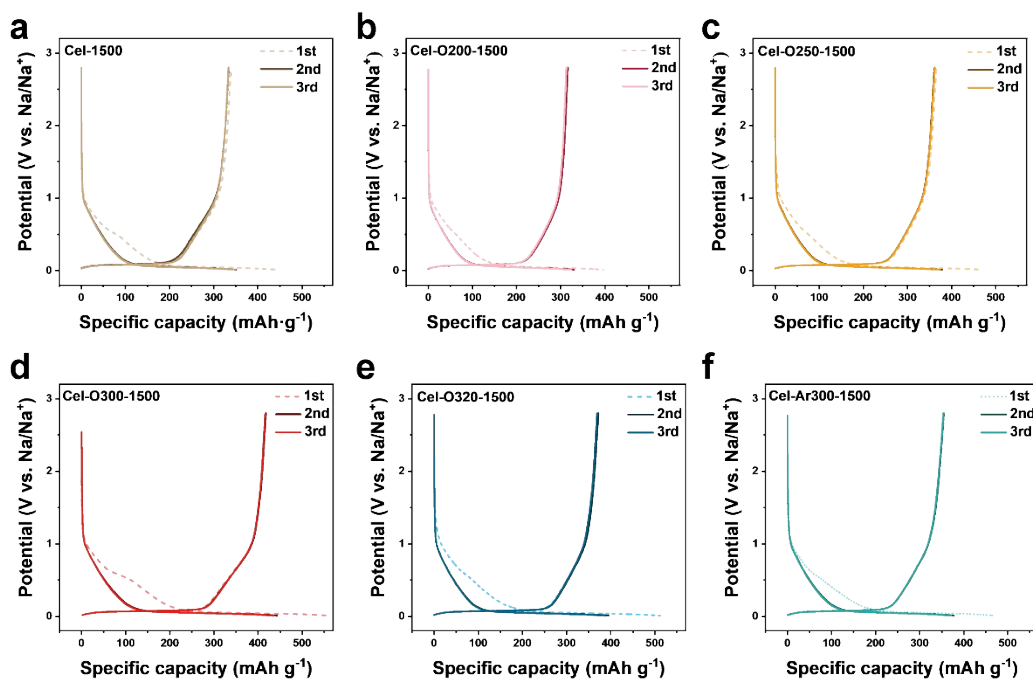


Fig. S8. GCD profiles of Cel-1500, Cel-Ox-1500 and Cel-Ar300-1500 during the initial three cycles.

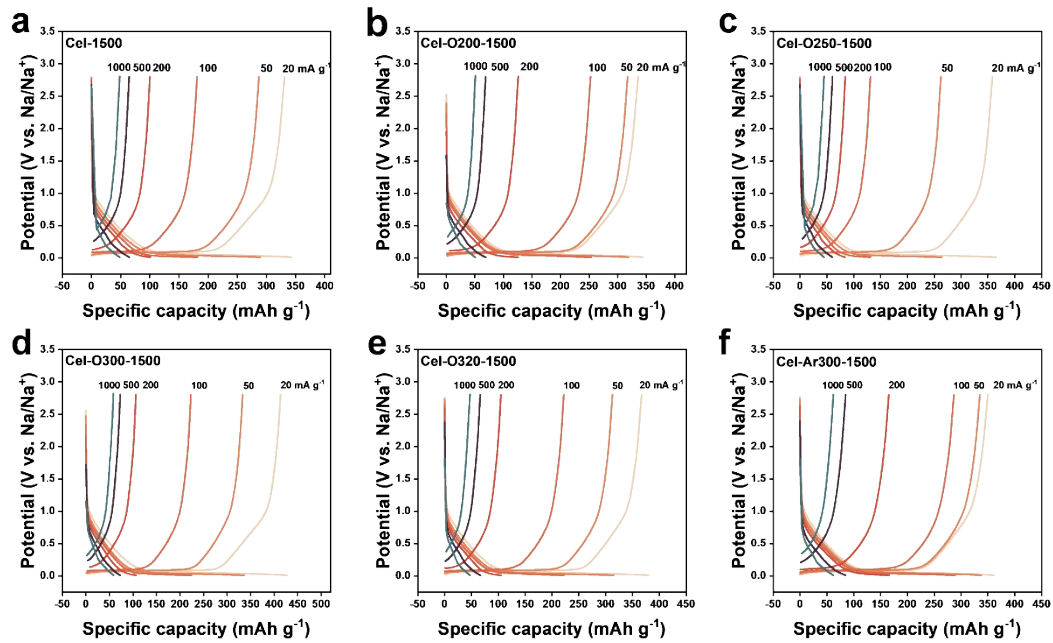


Fig. S9. GCD profiles of Cel-1500, Cel-Ox-1500 and Cel-Ar300-1500 at different current density in SIBs.

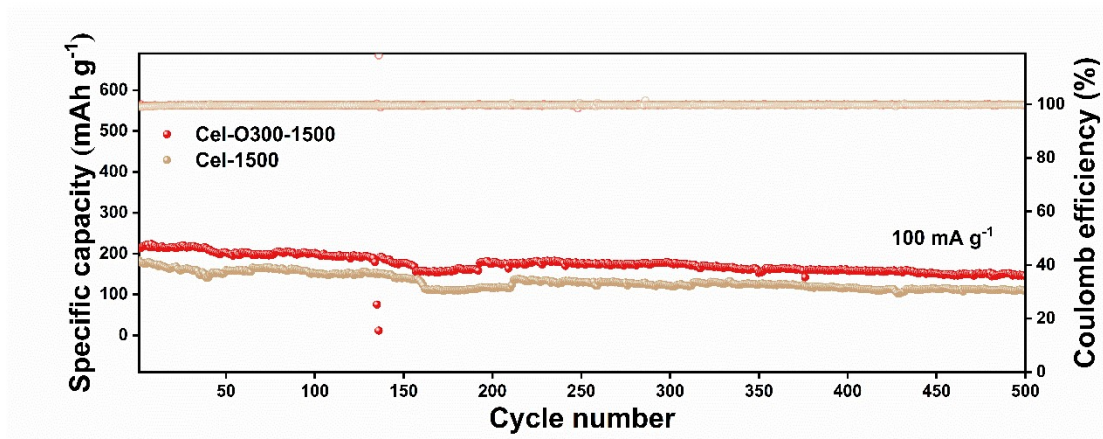


Fig. S10. long-term cycling performance and coulombic efficiency at 100 mA g⁻¹ of Cel-O300-1500 and Cel-1500.

Table S2. The electrochemical performance and plateau capacity ratio of cellulose-derived hard carbon samples measured at 20 mA g⁻¹.

Samples	Reversible Capacity (mAh g ⁻¹)	ICE (%)	Plateau Capacity (mAh g ⁻¹)	The proportion of the Plateau capacity
Cel-1500	342.5	76.6	235.2	0.687
Cel-O200-1500	344.4	71.9	238.8	0.693
Cel-O250-1500	365.6	77.9	262.8	0.719
Cel-O300-1500	426.8	74.8	307.5	0.721
Cel-O320-1500	380.2	72.4	275.5	0.725
Cel-Ar300-1500	361.0	75.6	246.1	0.682

Table S3. Kinetic fitting parameters and calculated D according to EIS test.

Samples	Re (Ω)	Rf (Ω)	Rct (Ω)	σ (Ω s ^{-1/2})	D (cm ² s ⁻¹)
Cel-1500	4.886	5.824	76.81	933.33	3.18478E-12
Cel-Ar300-1500	6.102	4.831	33.44	782.91	4.52612E-12
Cel-O300-1500	4.836	2.143	41.07	1135.99	2.14982E-12

Table S4. Summary of reported pre-oxidation-related and cellulose-derived hard carbon anodes for SIBs.

Samples	Pyrolysis temperature (°C)	S _{BET} (m ² g ⁻¹)	d (002) (nm)	Reversible capacity (mAh g ⁻¹)	ICE (%)	Ref.
Phenolic Resin (PFHC-20)	1100	35.3	0.389	334.3 (20 mA g ⁻¹)	84.7	1
Bituminous coal (RC-GO-1200)	1200	8.4	0.380	274.2 (30 mA g ⁻¹)	74.8	2

Esterified starch (H300-1100)	1100	2.9	0.387	369.8 (20 mA g ⁻¹)	82.5	3
D-glucose powder (G-220-1300)	1300	6.0	0.364	308.0 (25 mA g ⁻¹)	84.0	4
Asphalt (HC-325-9)	1100	367.9	0.389	286.9 (10 mA g ⁻¹)	83.6	5
Cotton	1300	38.0	0.410	260.0 (20 mA g ⁻¹)	72.0	6
Cellulose nanofibers	1000	377.0	—	255.0 (40 mA g ⁻¹)	58.8	7
Microcrystalline cellulose	1600	<10	0.376	310.0 (37.2 mA g ⁻¹)	84.0	8
Filter paper (DPC-A)	800	761.0	—	289.0 (20 mA g ⁻¹)	81.0	9
Nanocellulose (Spinifex derived)	1000	154.0	0.390	386.0 (20 mA g ⁻¹)	50.0	10
Cellulose nanocrystals (CNCs)	1000	145.6	0.390	340.0 (100 mA g ⁻¹)	—	11
Microcrystalline cellulose	1400	5.0	0.373	310.0 (37.2 mA g ⁻¹)	82.7	12
Cellulose powder derived from cotton linter	1300	506.0	0.389	351.0 (25 mA g ⁻¹)	68.0	13
Filter Paper (MV-HC)	1100	379.0	—	308.0 (20 mA g ⁻¹)	68.0	14
Microcrystalline cellulose (BHC-CO ₂)	1000	96.0	0.350	293.5 (50 mA g ⁻¹)	39.5	15
Nanocellulose powder	1200	—	—	250.0 (100 mA g ⁻¹)	—	16
Microcrystalline cellulose (HC-88.5)	1300	281.8	0.404	322.0 (50 mA g ⁻¹)	—	17
Microcrystalline cellulose (MCC-1400)	1400	5.8	0.388	343.3 (20 mA · g ⁻¹)	87.3	18
Cellulose fiber (HC-Cell)	1300	5.1	0.381	332.0 (0.2 C)	83.0	19
cellulose and lignin mixed at amass ratio of	1400	53.0	0.384	259.0 (100 mA g ⁻¹)	—	20

1:1 (CL-HC)						
bacterial cellulose (T-SBC-1300)	1300	51.2	0.390	239.0 (30 mA g ⁻¹)	56.9	²¹
α-cellulose (Cel-O300-1500)	1500	8.7	0.383	426.8 (20 mA g⁻¹)	74.8	This work

Reference

- 1 G. Zhang, L. Zhang, Q. Ren, L. Yan, F. Zhang, W. Lv and Z. Shi, *ACS Appl. Mater. Interfaces*, 2021, **13**, 31650–31659.
- 2 Z. Lou, H. Wang, D. Wu, F. Sun, J. Gao, X. Lai and G. Zhao, *Fuel*, 2022, **310**, 122072.
- 3 M. Song, Z. Yi, R. Xu, J. Chen, J. Cheng, Z. Wang, Q. Liu, Q. Guo, L. Xie and C. Chen, *Energy Storage Materials*, 2022, **51**, 620–629.
- 4 Z. V. Bobyleva, O. A. Drozhzhin, A. M. Alekseeva, K. A. Dosaev, G. S. Peters, G. P. Lakienko, T. I. Perfil'yeva, N. A. Sobolev, K. I. Maslakov, S. V. Savilov, A. M. Abakumov and E. V. Antipov, *ACS Appl. Energy Mater.*, 2023, **6**, 181–190.
- 5 R. Xu, Z. Yi, M. Song, J. Chen, X. Wei, F. Su, L. Dai, G. Sun, F. Yang, L. Xie and C.-M. Chen, *Carbon*, 2023, **206**, 94–104.
- 6 Y. Li, Y.-S. Hu, M.-M. Titirici, L. Chen and X. Huang, *Adv. Energy Mater.*, 2016, **6**, 1600659.
- 7 W. Luo, J. Schardt, C. Bommier, B. Wang, J. Razink, J. Simonsen and X. Ji, *J. Mater. Chem. A*, 2013, **1**, 10662.
- 8 V. Simone, A. Boulineau, A. de Geyer, D. Rouchon, L. Simonin and S. Martinet, *Journal of Energy Chemistry*, 2016, **25**, 761–768.
- 9 P. Zheng, T. Liu and S. Guo, *Sci Rep*, 2016, **6**, 35620.
- 10 R. R. Gaddam, E. Jiang, N. Amiralian, P. K. Annamalai, D. J. Martin, N. A. Kumar and X. S. Zhao, *Sustainable Energy Fuels*, 2017, **1**, 1090–1097.
- 11 H. Zhu, F. Shen, W. Luo, S. Zhu, M. Zhao, B. Natarajan, J. Dai, L. Zhou, X. Ji, R. S. Yassar, T. Li and L. Hu, *Nano Energy*, 2017, **33**, 37–44.
- 12 C. M. Ghimbeu, *Nano Energy*.
- 13 H. Yamamoto, S. Muratsubaki, K. Kubota, M. Fukunishi, H. Watanabe, J. Kim and S. Komaba, *J. Mater. Chem. A*, 2018, **6**, 16844–16848.
- 14 Z. Li, Y. Chen, Z. Jian, H. Jiang, J. J. Razink, W. F. Stickle, J. C. Neuefeind and X. Ji, *Chem. Mater.*, 2018, **30**, 4536–4542.
- 15 H. Wang, F. Sun, Z. Qu, K. Wang, L. Wang, X. Pi, J. Gao and G. Zhao, *ACS Sustainable Chem. Eng.*, 2019, **7**, 18554–18565.
- 16 L. Li, D. Chen, S. Su and B. Zeng, *mat express*, 2020, **10**, 1685–1691.
- 17 S. Alvin, C. Chandra and J. Kim, *Chemical Engineering Journal*, 2021, **411**, 128490.
- 18 X.-S. Wu, X.-L. Dong, B.-Y. Wang, J.-L. Xia and W.-C. Li, *Renewable Energy*, 2022, **189**, 630–638.
- 19 J. Conder, C. Villevieille, J.-M. Le Meins and C. Matei Ghimbeu, *ACS Appl. Energy Mater.*, 2022, **5**, 12373–12387.
- 20 Q. Jin, L. Tao, Y. Feng, D. Xia, G. A. Spiering, A. Hu, R. Moore, F. Lin and H. Huang, *ACS Sustainable Chem. Eng.*, 2023, **11**, 536–546.
- 21 F. Wang, T. Zhang and F. Ran, *Electrochimica Acta*, 2023, **441**, 141770.