Engineering of TiO₂ Anode toward a Record High Initial Coulombic Efficiency Enabling High-Performance Low-Temperature Na-ion Hybrid Capacitors

Meiling Kang,^{a, b} Yingying Wu,^a Xin Huang,^a Kaiqiang Zhou,^{a, b} Zhigao Huang^{a, b} and

Zhengsheng Hong*, a, b

^aFujian Provincial Key Laboratory of Quantum Manipulation and New Energy Materials,

College of Physics and Energy, Fujian Normal University, Fuzhou, Fujian 350117, China.

^bFujian Provincial Collaborative Innovation Center for Optoelectronic Semiconductors and

Efficient Devices, Xiamen, 361005, China

*Corresponding Authors: E-mail: winter0514@163.com



Fig. S1. TG curves of TiO_2 (a) and TiO_2 -S (b).



Fig. S2. (a) Nitrogen adsorption-desorption curves of TiO_2 and TiO_2 -S, (b) the corresponding BJH pore size distribution.



Fig. S3. SEM images of (a, b) the as-made product, (c, d) TiO₂ (e) EDS pattern of TiO₂-S.



Fig. S4. a) Charge-discharge profiles for TiO_2 in ether and ester under 0.05 A/g. b) Cycling stability of the different electrodes in different electrolytes.



Fig. S5. CV curves of TiO_2 -S electrode in ether electrolyte under high sweep rates.



Fig. S6. EIS plots for a) different electrodes in ether and b) the TiO_2 electrode in different electrolytes.



Fig. S7. SEM images for TiO_2 -S electrode in (a, b) ether electrolyte and (c, d) in ester after the first discharge. The inset in b is the enlarged SEM image.



Fig. S8. The elements mapping (a) as well as correspinding full area EDS (b) of TiO_2 -S electrodes after first discharge in ester.

a



Fig. S9. The elements mapping (a) as well as correspinding full area EDS (b) of TiO_2 -S electrodes after first discharge in ether.

Table S1 The various element mass percent in titanium dioxide nanosheets with sulfur doing in ester electrodes after first discharge.

Element	Ti	0	С	Na	F	Р	S
Ether (%)	10.1	47.03	20.37	20.9	1.16	0.29	0.16
Ester (%)	10.22	37.99	35.12	14.07	2.01	0.41	0.18



Fig. S10. The electrochemical performance of NVP cathode evaluated by half-cells within different electrolytes: a) rate capability combining with b) charge/discharge profiles at the selective cycles at 0.1 A/g.

Anode//Cathode	Potential	Energy	Power	Cycling life	Reference
	range (V)	density	density		
		(Wh/kg)	(W/kg)		
V ₂ O ₅ -CNT//AC	0-2.8	38	140	≈80% at 60C after	
				900 cycles	
MWTOG//AC	0.01-3	25.8	1357	≈90% at 10C after	
				10000 cycles	
Na ₃ V ₂ (PO ₄) ₃ @C//AC	0-3	118	96	≈95% at 1.1 mA cm ⁻	1
				² after 10 000 cycles	
Na ₂ Ti ₉ O ₁₉ //PC	0-2.5	54	687	\approx 75% at 2 A g ⁻¹ after	2
				2000 cycles	
Nb ₂ O ₅ @C/rGO//AC	1-4.3	76	80	$\approx 100\%$ at 1 A g ⁻¹	3
				after 3000 cycles	
SCN-A//SCN-A	0-4	112	67	≈85% at 5 A g ⁻¹ after	4
				3000 cycles	
Na-TNTs//AC	0-3	34	889	≈90% at 0.25 A g ⁻¹	5
				after 1000 cycles	
NaTi ₂ O ₄ (OH) ₂ //PC	0-3	65	500	\approx 93% at 1 A g ⁻¹ after	6
				3000 cycles	
Nb ₂ O ₅ //PSC	1-3	43.2	5760	≈80% at 1.2 A g ⁻¹	7
				after 3000 cycles	
N-TiO ₂ //AC	1-4	80.3	500	$\approx 80\%$ at 1 A g ⁻¹ after	
				6500 cycles	
TiO ₂ -S//Na ₂ V ₃ (PO4) ₃	0.01-3.5	158	1075	$\approx 83\%$ at 1 A g ⁻¹ after this work	
				1000 cycles	

 Table S2 Comparison of representative electrochemical energy storage devices.

REFERENCES

- 1 R. Thangavel, K. Kaliyappan, K. Kang, X. Sun and Y. S. Lee, *Adv. Energy Mater.*, 2016, 6, 1–9.
- 2 S. S. M. Bhat, B. Babu, M. Feygenson, J. C. Neuefeind and M. M. Shaijumon, *ACS Appl. Mater. Interfaces*, 2018, **10**, 437–447.
- 3 E. Lim, J. Changshin, M. S. Kim, M. H. Kim, J. Chun, H. Kim, J. Park, K. C. Roh, K. Kang, S. Yoon and J. Lee, *Adv. Funct. Mater.*, 2016, **26**, 3711–3719.
- 4 H. Wang, D. Mitlin, J. Ding, Z. Li and K. Cui, J. Mater. Chem. A, 2016, 4, 5149–5158.
- 5 J. Yin, L. Qi and H. Wang, ACS Appl. Mater. Interfaces, 2012, 4, 2762–2768.
- 6 B. Babu and M. M. Shaijumon, J. Power Sources, 2017, **353**, 85–94.
- H. Li, Y. Zhu, S. Dong, L. Shen, Z. Chen, X. Zhang and G. Yu, *Chem. Mater.*, 2016, 28, 5753–5760.