Electronic supplementary information

Strong (001) facet-induced growth of multi-hierarchical tremella-like Sndoped V_2O_5 for high-performance potassium-ion batteries

Lidong Xing¹, Qiyao Yu^{*,2}, Yanping Bao^{*,1}, Jianhua Chu¹, Kun Han¹, Shaokun Chong³, Cheng-Yen Lao⁴, Feili Lai⁵, Ping Li¹, Kai Xi⁴, Wei (Alex) Wang^{*,6}

¹State Key Laboratory of Advanced Metallurgy, University of Science and Technology Beijing, Beijing 100083, China.

²Institute of Advanced Structure Technology, Beijing Institute of Technology, Beijing 100081, China ³John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge, MA 02138, USA

⁴Department of Materials Science and Metallurgy, University of Cambridge, Cambridge CB3 0FS, UK.

⁵Hefei National Laboratory for Physical Science at the Microscale, University of Science and Technology of China, Hefei, Anhui 230026, China.

⁶Beijing Key Laboratory of Bio-inspired Energy Materials and Devices, School of Space and Environment, Beihang University, Beijing, 100191 PR, China



Fig. S1 Morphological evolution of SDVO at different times: (a) 6 h, (b) 12 h, (c) 24 h.



Fig. S2 HRTEM images of tremella-like pc-SDVO before calcination.



Fig. S3 (a) FESEM, (b-d) TEM and (e) HRTEM images of V_2O_5 NPs after calcination.



Fig. S4 (a) FESEM image and (b) photograph of tremella-like pc-SDVO before calcination. (c) FESEM image and (d) photograph of tremella-like SDVO after calcination. (e) FESEM image and (f) photograph of V_2O_5 NPs after calcination.



Fig. S5 (a-d) HRTEM images of tremella-like crystallized SDVO after calcination.

The formation of mesoporous can be attributed to the fact that the V_2O_5 nanosheets reacted with excessive Sn^{4+} cations through selective cation exchange.^{1, 2}



Fig. S6 (a) HRTEM image and (b) interplanar spacing measurement chart of tremella-like crystallized SDVO.



Fig. S7 (a) STEM and (b-d) the corresponding elemental mapping images of tremella-like pc-SDVO before calcination.



Fig. S8 The EDS spectrum of tremella-like SDVO.



Fig. S9 (a) Nitrogen adsorption-desorption isotherms of V_2O_5 NPs. (b) Pore size distribution of the V_2O_5 NPs.



Fig. S10 XRD patterns of tremella-like pc-SDVO before calcination.



Fig. S11 XPS spectra of (a) V 2p and (b) O 1s in V_2O_5 NPs.



Fig. S12 XPS spectra of (a) V 2p, (b) O 1s, (c) Sn 3d in pc-SDVO before calcination.



Fig. S13 XPS survey spectrum of crystallized SDVO.



Fig. S14 Selected charge/discharge voltage profiles at current density of 0.1 A g⁻¹ with (a-b) 1# and (c) 2# electrolyte.



Fig. S15 The cycle performance of V_2O_5 NPs and pc-SDVO with 2# electrolyte.



Fig. S16 (a) FESEM images and (b) XRD pattern of tremella-like V_2O_5 without Sn doping. (c) Cycle performance and the corresponding Coulomb efficiency of tremella-like V_2O_5 at 0.1 A g⁻¹.

In a typical synthesis of tremella-like V₂O₅, 0.28 g of ammonium metavanadate and 0.50 g of citric acid were dissolved in a mixed solvent of ethanediol (30 mL) and deionized water (10 mL); this mixture resulted in a light yellow solution. This solution was stirred for 5 h at 60 °C until it turned atrovirens, then it was transferred to a 50 mL Teflon autoclave, and kept at 180 °C for 24 h. After naturally cooling to an ambient temperature, the precipitates were mildly and repeatedly washed with pure alcohol and deionized water, and dried at 80 °C for 6 h in air. The samples were obtained after heating at 400 °C for 2 h in air.³



Fig. S17 CV curves from 1st to 3rd cycle at a scan rate of 0.1 mV s⁻¹ with 1# electrolyte.



Fig. S18 The rate performance of pc-SDVO, V_2O_5 NPs and tremella-like V_2O_5 with 2# electrolyte.



Fig. S19 (a) SEM and (b)TEM images of SDVO electrodes after 3000 cycles.



Fig. S20 The electrochemical reaction dynamics of SDVO with 2# electrolyte. (a) CV profiles and (b) the percentages of pseudocapacitive contributions at different scan rates. (c) The plots of log (peak current) *vs.* log (scan rates). (d) Relationship between $i(V)/v^{1/2} vs. v^{1/2}$ used for calculating constants k_1 and k_2 .

When the *b*-value is close to 0.5, the electrochemical behavior is dominated by the ionic diffusion process, while the *b*-value close to 1.0, the capacitive process was dominated. When the b-value is between 0.5 and 1.0, it is suggested the mixed mechanisms controlled during the energy storage process. The ratio of two mechanism in quantity could be calculated using the formula of $i = k_1v + k_2v^{1/2}$ and the corresponding results was shown in Fig. S20b[†]. In the formula, k_1v and $k_2v^{1/2}$ describe the current contributions from the capacitive and diffusion processes, respectively. According to the above formula, we could get a straight line with a slope of k_1 , and intercept of k_2 shown in Fig. S20d[†].



Fig. S21 EIS plots of the tremella-like SDVO with different electrolyte at different cycles.

Element	Atom/%	Weight ratio/%
V	55.58	74.4
0	41.91	17.6
Sn	2.51	7.9
Totals	100	100

Table S1. The element content of tremella-like SDVO via EDS mapping analysis.

Element	Atom ratio	
V	22	
Sn	1	

 Table S2. The weight ratio of tremella-like SDVO.

Sample	Electronic conductivity (S m ⁻¹)		
V ₂ O ₅	1.9×10 ⁷		
SDVO	5.72×10 ⁷		

Table S3. A comparison of the electronic conductivity of different materials.

Crystal facet	Surface energy		
	V ₂ O ₅	SDVO	
(001)	0.00257	0.02489	
(110)	0.03436	0.05683	
(100)	0.03539	0.05883	
(010)	0.02455	0.04554	

Table S4. The first-principles calculations on the surface energy of V_2O_5 and SDVO.

Sample	Current density	Capacity	Cycle	Referenc
	(A g ⁻¹)	(mAh g ⁻¹)	number	e
<u>CDVO</u>	0.1	308	150	This work
SDVO	0.5	188	3000	I nis work
V ₂ O ₃ @PNCNFs	0.05	240	500	4
SnO ₂ -G-C	0.1	285	60	5
G@C@SnO ₂	0.05	148	85	6
K ₂ BPDC@GR	0.05	170	100	7
Vitamin K3@GNT	0.1	222	100	8
KMnF-LE	0.1	138	100	9
Hard Carbon	0.03	216	100	10
S, O-Codoped Hard Carbon	0.05	226	100	11
N-doped Graphene	0.1	210	100	12

Table S5. Electrochemical performance survey of some reported anode materials of KIBs.

References

- L. Li, S. Peng, H. B. Wu, L. Yu, S. Madhavi and X. W. D. Lou, *Adv. Energy Mater.*, 2015, 5, 1500753.
- L. Zhang, M. Yang, S. Zhang, Z. Wu, A. Amini, Y. Zhang, D. Wang, S. Bao, Z. Lu, N. Wang and C. Cheng, *Sci Rep*, 2016, 6, 33597.
- C. Niu, M. Huang, P. Wang, J. Meng, X. Liu, X. Wang, K. Zhao, Y. Yu, Y. Wu, C. Lin and L. Mai, *Nano Research*, 2015, 9, 128-138.
- 4. T. Jin, H. Li, Y. Li, L. Jiao and J. Chen, *Nano Energy*, 2018, **50**, 462-467.
- 5. Z. Huang, Z. Chen, S. Ding, C. Chen and M. Zhang, *Nanotechnology*, 2018, **29**, 375702.
- Q. Wu, Q. Shao, Q. Li, Q. Duan, Y. Li and H. G. Wang, ACS Appl. Mater. Interfaces, 2018, 10, 15642-15651.
- C. Li, Q. Deng, H. Tan, C. Wang, C. Fan, J. Pei, B. Cao, Z. Wang and J. Li, ACS Appl. Mater. Interfaces, 2017, 9, 27414-27420.
- Q. Xue, D. Li, Y. Huang, X. Zhang, Y. Ye, E. Fan, L. Li, F. Wu and R. Chen, *J. Mater. Chem. A*, 2018, 6, 12559-12564.
- Z. Liu, P. Li, G. Suo, S. Gong, W. Wang, C.-Y. Lao, Y. Xie, H. Guo, Q. Yu, W. Zhao, K. Han,
 Q. Wang, M. Qin, K. Xi and X. Qu, *Energy Environ. Sci.*, 2018, 11, 3033-3042.
- 10. Z. Jian, Z. Xing, C. Bommier, Z. Li and X. Ji, Adv. Energy Mater., 2016, 6, 1501874.
- M. Chen, W. Wang, X. Liang, S. Gong, J. Liu, Q. Wang, S. Guo and H. Yang, *Adv. Energy Mater.*, 2018, 8, 1800171.
- 12. K. Share, A. P. Cohn, R. Carter, B. Rogers and C. L. Pint, ACS Nano, 2016, 10, 9738.