

Bio-inspired high-performance solid-state supercapacitors with electrolyte, separator, binder and electrodes entirely from *kelp*

Juan Zeng, Lu Wei and Xin Guo

Laboratory of Solid State Ionics, School of Materials Science and Engineering, Huazhong University of Science and Technology, Wuhan 430074, P. R. China

Table S1. Property parameters of the commercial activated carbon TF-B520

Density	0.32-0.40 g cm ⁻³
Particle size	5(±1) μm
Ash content	<0.07%
pH value	6.0-7.0
Specific surface area	2000 (±100) m ² g ⁻¹
Pore volume	1.0-1.2 cm ³ g ⁻¹
Pore size	2.0-2.2 nm
Carbon content	>95%
Metal ions from ICP	<1000 ppm

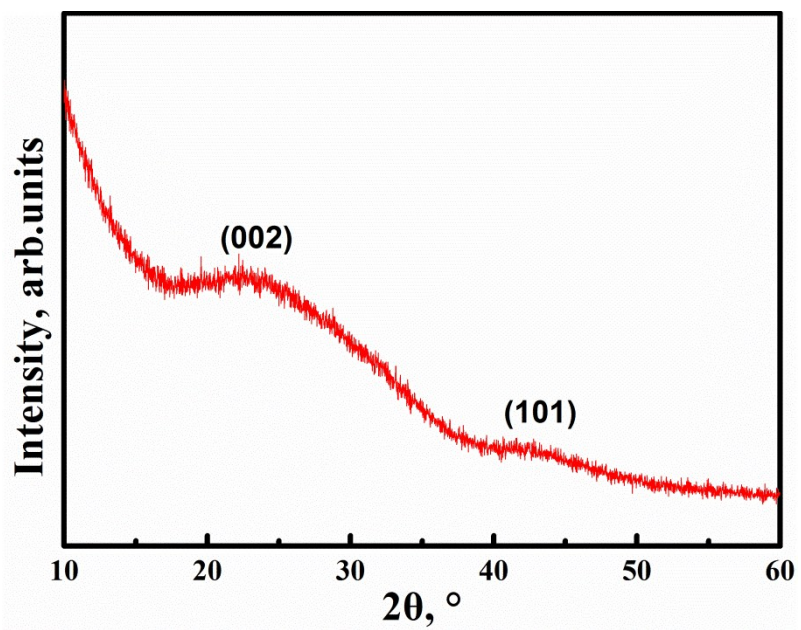


Fig. S1 XRD pattern of the *kelp*-derived activated carbon (KAC).

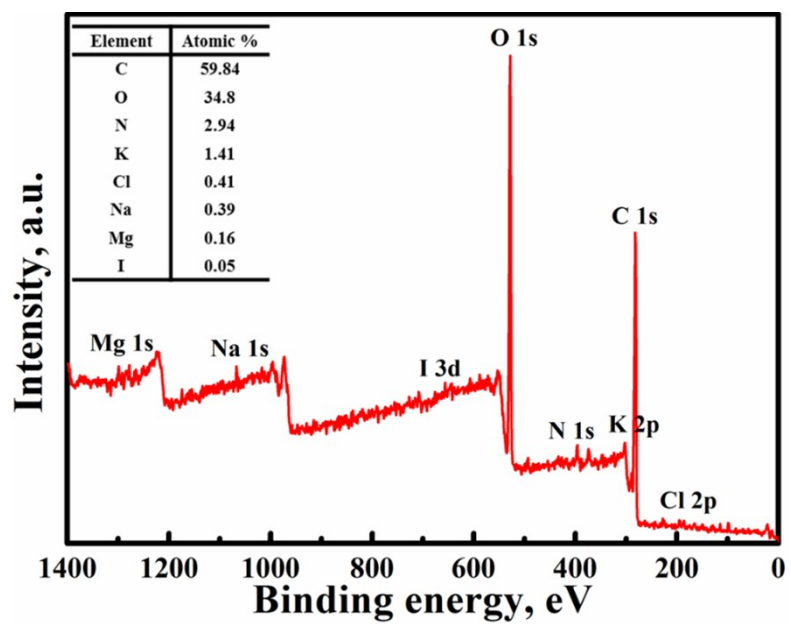


Fig. S2 XPS spectrum with all elements detected in *kelp*, the inset shows the atomic contents of each element.

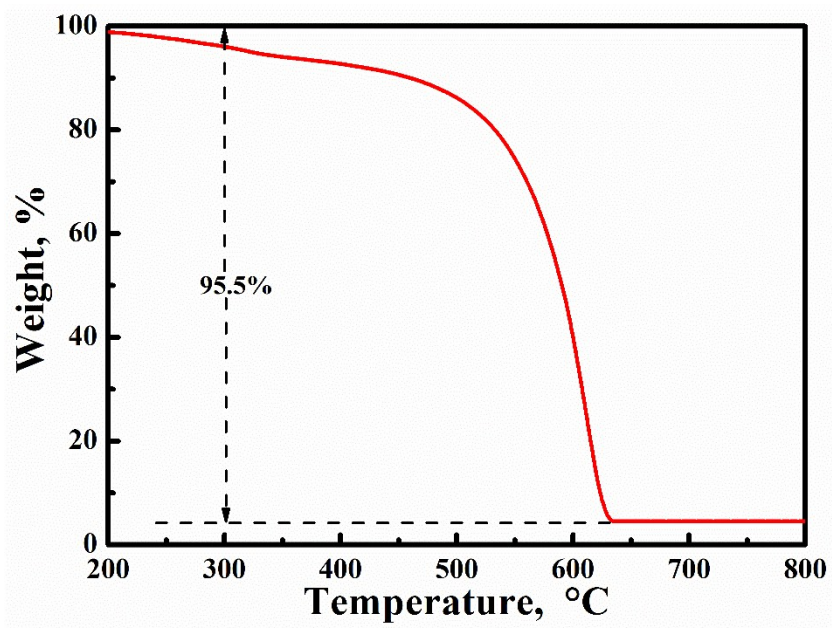


Fig. S3 Thermogravimetric curve of KAC in air.

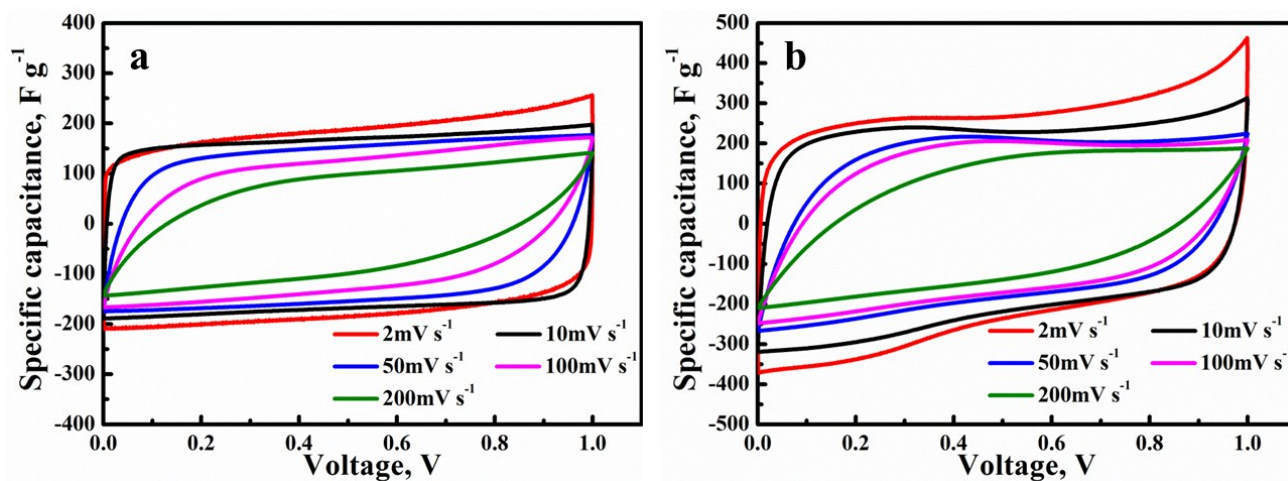


Fig. S4 CV curves of (a) supercapacitor based on commercial activated carbon (TF-B520) electrodes, Na-alginate binder, Na-alginate separator and Na-alginate hydrogel electrolyte, and (b) supercapacitor with KAC electrodes, PTFE binder, commercial polypropylene separator and 1 M H₂SO₄ aqueous electrolyte.

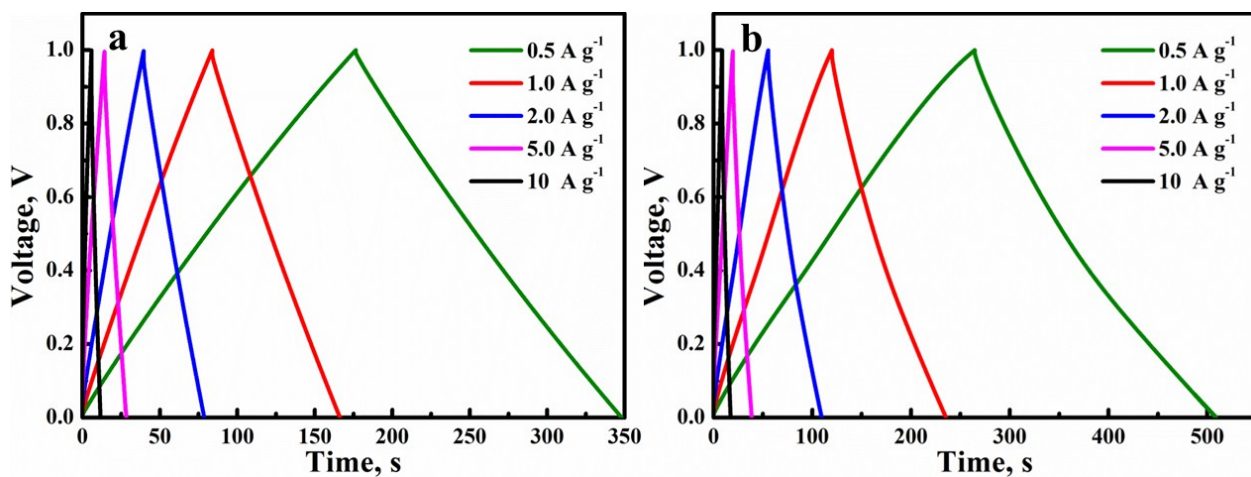


Fig. S5 C-D curves of (a) supercapacitor based on commercial activated carbon (TF-B520) electrodes, Na-alginate binder, Na-alginate separator and Na-alginate hydrogel electrolyte, and (b) supercapacitor with KAC electrodes, PTFE binder, commercial polypropylene separator and 1 M H_2SO_4 aqueous electrolyte.

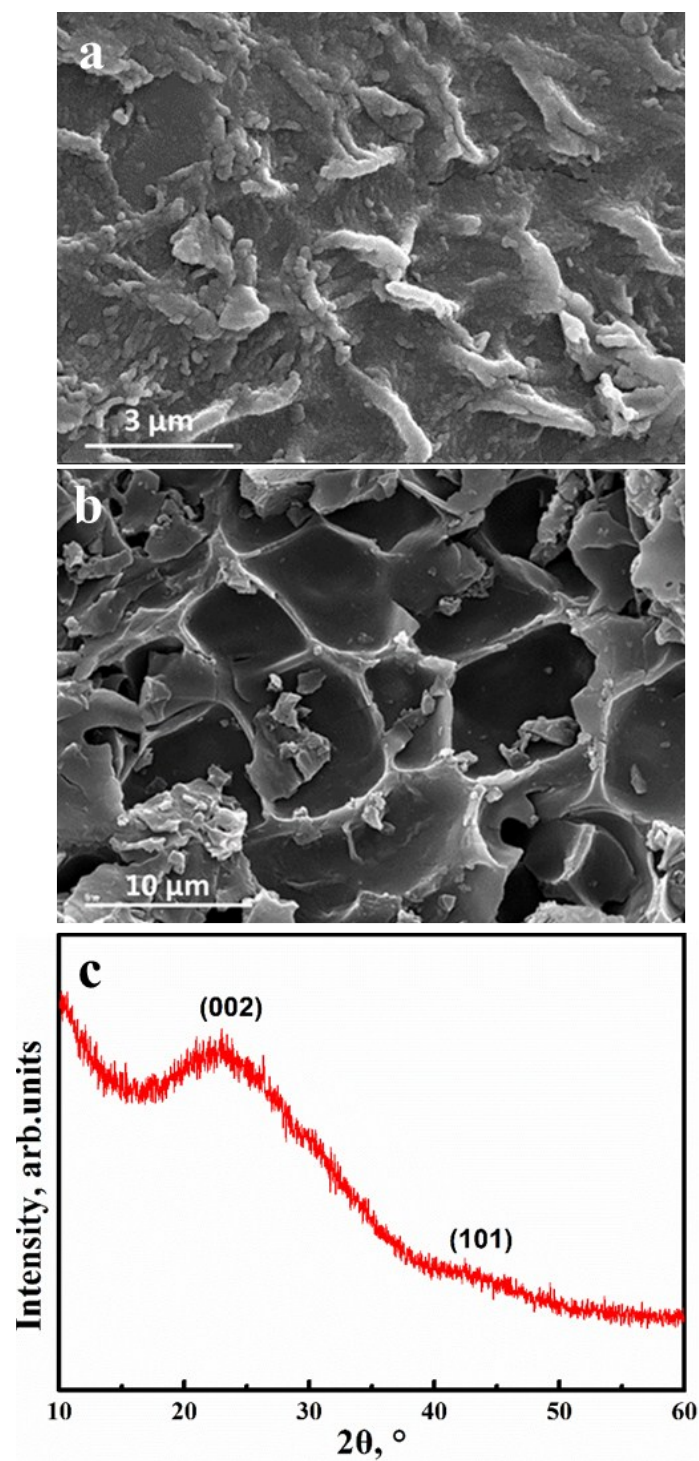


Fig. S6 (a, b) SEM images and (c) XRD pattern of KAC electrode material after charge-discharge cycling tests.

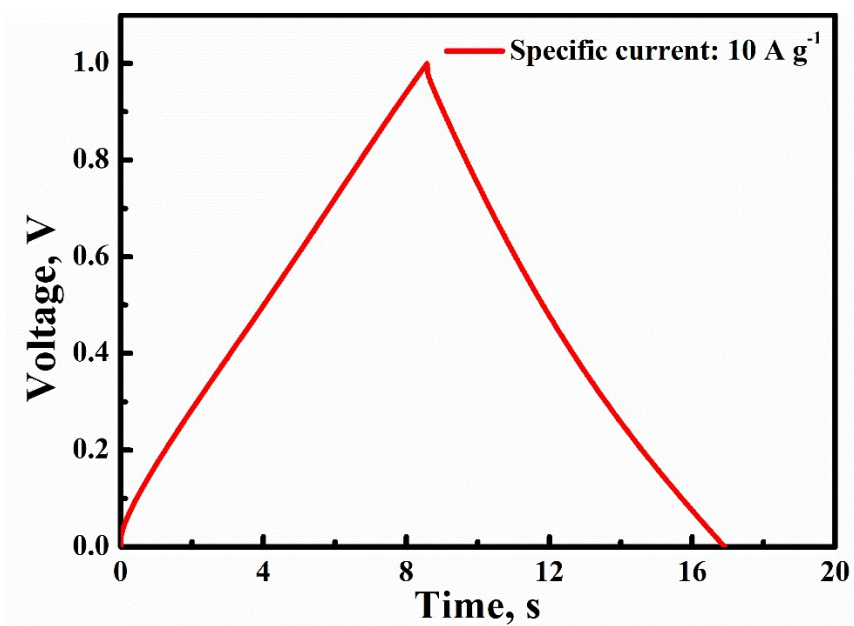


Fig. S7 C-D curve of the “all-kelp” solid-state supercapacitor re-tested at 60 °C after 10,000 C-D cycles at 10 A g⁻¹.

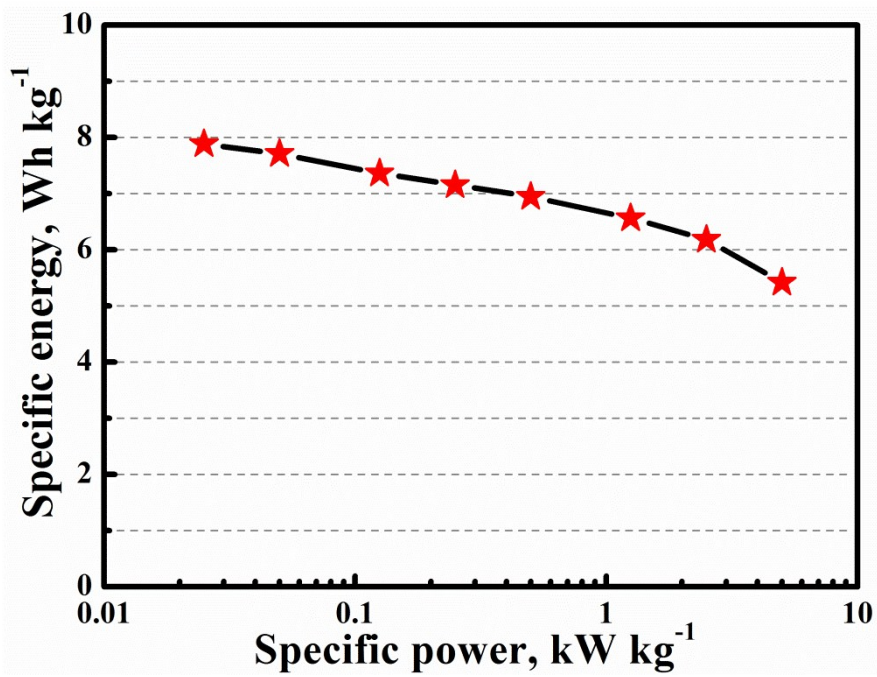


Fig. S8 Ragone plot of the "all-kelp" solid-state supercapacitor.

Table S2 Carbon electrode materials for all-solid-state supercapacitors

Electrode material	Doped	SSA (m ² g ⁻¹)	Electrolyte	Capacitance (F g ⁻¹)	Reference
<i>Kelp</i> -derived activated carbon	—	4425	Na-alginate gel	227	This work
Graphene hydrogel film	—	414	PVA-H ₂ SO ₄ gel	186	1
Carbon nanotubes	—	456	[EMIM][NTf ₂]-silica gel	135	2
Graphene-cellulose paper	—	—	PVA-H ₂ SO ₄ gel	120	3
3D graphene	—	1661	PVA-H ₂ SO ₄ gel	80	4
Activated carbon fibers	—	1476	PVA-H ₃ PO ₄ gel	43.8	5
Activated carbon cloth	—	61.2	PVA-H ₂ SO ₄ gel	0.00155	6
Carbon nanofibers	<i>N</i> -	312	PVA-H ₂ SO ₄ gel	175	7
Graphene aerogel	<i>N,B</i> -	249	PVA-H ₂ SO ₄ gel	62	8

References

1. Y. X. Xu, Z. Y. Lin, X. Q. Huang, Y. Liu, Y. Huang and X. F. Duan, *ACS Nano*, 2013, **7**, 4042–4049.
2. Y. J. Kang, H. Chung, C. H. Han and W. Kim, *Nanotechnology*, 2012, **23**, 065401.
3. Z. Weng, Y. Su, D. W. Wang, F. Li, J. H. Du and H. M. Cheng, *Adv. Energy Mater.*, 2011, **1**, 917–922.
4. A. Ramadoss, K. Y. Yoon, M. J. Kwak, S. I. Kim, S. T. Ryu and J. H. Jang, *J. Power Sources*, 2017, **337**, 159–165.
5. W. J. Ma, S. H. Chen, S. Y. Yang, W. P. Chen, W. Weng and M. F. Zhu, *ACS Appl. Mater. Interfaces*, 2016, **8**, 14622–14627.
6. G. M. Wang, H. Y. Wang, X. H. Lu, Y. C. Ling, M. H. Yu, T. Zhai, Y. X. Tong and Y. Li, *Adv. Mater.*, 2014, **26**, 2676–2682.
7. L. F. Chen, Z. H. Huang, H. W. Liang, W. T. Yao, Z. Y. Yu and S. H. Yu, *Energy Environ. Sci.*, 2013, **6**, 3331–3338.
8. Z. S. Wu, A. Winter, L. Chen, Y. Sun, A. Turchanin, X. Feng and K. Müllen, *Adv. Mater.*, 2012, **24**, 5130–5135.

Table S3 Comparison of the electrochemical performance of advanced all-solid-state supercapacitors with carbon-based materials reported in literatures. Here (E) represents the calculation method based on electrode level and (D) represents calculation method based on device level.

Electrode material	Gravimetric capacitance (F g ⁻¹)	Volumetric capacitance (F cm ⁻³)	Volumetric energy density (Wh L ⁻¹)	Cycling retention	Reference
<i>Kelp</i> -derived AC	227	72.3 (E) 18.1 (D)	2.5	97% (10,000 cycles)	This work
Graphene hydrogel film	186	31 (E)	1.07	97.5% (10,000 cycles)	1
CNT fiber//CNT sheet	59	32.1 (E)	1.11	—	2
AC fiber	43.8	27.6 (D)	2.5	90.4% (10,000 cycles)	3
Carbon nanofiber	84	2.1 (D)	2.5	96% (10,000 cycles)	4
MWCNT/carbon microfiber	11.1	14.1 (E)	0.14	94% (1000 cycles)	5
AWC//WC@MnO ₂	35.6	14.4 (D)	6.4	93% (10,000 cycles)	6
CNT/vanadium nitride	5.2	7.9 (D)	0.54	82% (10,000 cycles)	7
H-TiO ₂ @MnO ₂ //H-TiO ₂ @C	139.6	0.7 (D)	0.3	91.2% (5000 cycles)	8
AC//polypyrrole@MnO ₂	261	19.3 (D)	8.67	98.6% (1000 cycles)	9
Graphene//Co ₃ O ₄	117	2.1 (D)	0.62	84% (1000 cycles)	10

AC: activated carbon; CNT: carbon nanotube; MWCNT: multi-wall carbon nanotube; AWC: activated wood carbon; WC: wood carbon.

References

1. Y. X. Xu, Z. Y. Lin, X. Q. Huang, Y. Liu, Y. Huang and X. F. Duan, *ACS Nano*, 2013, **7**, 4042–4049.
2. X. L. Chen, L. B. Qiu, J. Ren, G. Z. Guan, H. J. Lin, Z. T. Zhang, P. N. Chen, Y. G. Wang and H. S. Peng, *Adv. Mater.*, 2013, **25**, 6436–6441.
3. W. J. Ma, S. H. Chen, S. Y. Yang, W. P. Chen, W. Weng and M. F. Zhu, *ACS Appl. Mater. Interfaces*, 2016, **8**, 14622–14627.
4. Y. M. Sun, R. B. Sills, X. L. Hu, Z. W. Seh, X. Xiao, H. H. Xu, W. Luo, H. Y. Jin, Y. Xin, T. Q. Li, Z. L. Zhang, J. Zhou, W. Cai, Y. H. Huang and Y. Cui, *Nano Lett.*, 2015, **15**, 3899–3906.
5. V. T. Le, H. Kim, A. Ghosh, J. Kim, J. Chang, Q. A. Vu, D. T. Pham, J. H. Lee, S. W. Kim and Y. H. Lee, *ACS Nano*, 2013, **7**, 5940–5947.
6. C. J. Chen, Y. Zhang, Y. J. Li, J. Q. Dai, J. W. Song, Y. G. Yao, Y. H. Gong, I. Kierzewski, J. Xie and L. B. Hu, *Energy Environ. Sci.*, 2017, **10**, 538–545.
7. X. Xiao, X. Peng, H. Y. Jin, T. Q. Li, C. C. Zhang, B. Gao, B. Hu, K. F. Huo and J. Zhou, *Adv. Mater.*, 2013, **25**, 5091–5097.
8. X. H. Lu, M. H. Yu, G. M. Wang, T. Zhai, S. L. Xie, Y. C. Ling, Y. X. Tong and Y. Li, *Adv. Mater.*, 2013, **25**, 267–272.
9. J. Y. Tao, N. S. Liu, L. Y. Li, J. Su and Y. H. Gao, *Nanoscale*, 2014, **6**, 2922–2928.
10. X. F. Wang, B. Liu, R. Liu, Q. F. Wang, X. J. Hou, D. Chen, R. M. Wang and G. Z. Shen, *Angew. Chem.*, 2014, **53**, 1849–1853.