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

Cypress Leaf-Like Sb as Anode Material for High-Performance Sodium-Ion Batteries

Hongshuai Hou, Mingjun Jing, Yan Zhang, Jun Chen, Zhaodong Huang and Xiaobo Ji*

College of Chemistry and Chemical Engineering, Central South University, Changsha, China. Fax: +86-731-88879616; Tel: +86-731-88879616; E-mail: xji.csu.edu@gmail.com

1. Experimental Sections

1.1 Materials

$SbCl_3$ (Shanghai Titan Scientific Co., Ltd.), ethanediol (Sinopharm Chemical Reagent Co., Ltd.), ethanol (Shanghai Titan Scientific Co., Ltd.), Mg powder (Shanghai Longxin technology development Co., Ltd.), hydrochloric acid (Sinopharm Chemical Reagent Co., Ltd.), $NaClO_4$ (Alfa Aesar), Na (Sigma-Aldrich), fluoroethylene carbonate (FEC, Sigma-Aldrich), propylene carbonate (Sigma-Aldrich) and carboxymethyl cellulose (CMC, Alfa Aesar).

1.2 Preparation of cypress leaf-like Sb (CL-Sb)

2.0 g of $SbCl_3$ was dissolved into 80 mL ethanediol, and then 0.4 g of commercial Mg powder was added under vigorous magnetic stirring. After 2 h, 120 mL diluted hydrochloric acid (1 mol L^{-1}) was added to remove the unreacted excess Mg. Vigorous stirring was maintained throughout all synthesis process. Finally, the product was collected by centrifugation and washed several times with deionized water and ethanol, followed by drying at 100 ºC under vacuum.

1.3 Characterization

Transmission electron microscope (TEM, JEM-2100F), scanning electron microscope (SEM, FEI Quanta 200) and X-ray diffraction (XRD, Rigaku D/max 2550 VB+ 18kW, Cu Kα radiation) were used to characterize the morphology and composition of samples.

1.4 Electrochemical tests

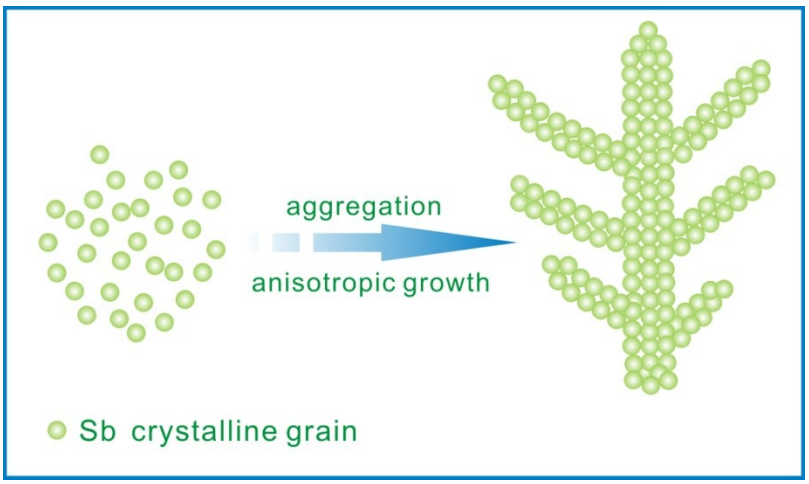
For electrochemical performance evaluation, coin-type half-cells were assembled in a glove box filled with Ar to investigate the sodium storage behaviors of as-prepared CL-Sb. A homogenous slurry was obtained through mixing the synthesized active materials with super P and carboxymethyl cellulose (70:15:15 in weight) in deionized water, and then it was painted on a copper foil. After solvent evaporation, the copper foil coated with active materials was cut, pressed and further dried at 100 °C under vacuum for 12 h. The tailored Cu foil coated with active materials was utilized as work electrode and metallic sodium was employed as the counter electrode, and Celgard 2400 was used as the separator. The electrolyte was a solution of 1 M $NaClO_4$ in propylene carbonate (PC) with 5% fluoroethylene carbonate (FEC) additive. Cyclic voltammetric measurements were performed on Solartron Analytical to examine the cathodic and anodic reaction using the above-mentioned cell in the voltage range of 2.0 - 0.01 V (vs. Na/Na^+) at a scan rate of 0.1 mV s^{-1}. Galvanostatic charge-discharge tests were carried out on Arbin battery cycler (BT2000) at suitable current densities between 0.01V and 2.0 V (vs. Na/Na^+) for both charge (Na extraction) and discharge (Na insertion) at room temperature. The electrochemical impedance measurements were performed on Solartron Analytical at an AC voltage of 5 mV amplitude in the 100 kHz to 0.01 Hz after 5 charge-discharge cycles. The mass loading of the active material is about 1.0-1.2 mg.

2. Formation process of CL-Sb

As reported, the leaf-like patterns can be formed only through anisotropic growth of crystalline grains.[1-3] Here, the anisotropy comes from the Sb particles themselves, becauce Sb inherently posesses strong crystalline anisotropy.[1] As is well known, Mg is a kind of very active metal and it is more active than metallic Sb. When Mg was added to the $SbCl_3$ solution, Sb^{3+} would be reduced to Sb, leading to the formation of a large number of fine Sb particles. Then, as illustrated in Scheme S1, the newly generated Sb particles collided and directionally aggregated resulting from the free diffusion and interaction among the Sb particles, and the large aggregated particles grew strictly along the preferred growth direction, resulting in the formation of the leaf-like crystal.

Table S1 Comparison of the electrochemical performances of the as-prepared CL-Sb with previously reported Sb-based anode materials for SIBs.

Materials	Initial reversible capacity (mAh g^{-1})	Cyclability (mAh g^{-1})	Current density (mA g^{-1})	References
Sn-Ge-Sb	833	662 after 50 cycles	85	4
$FeSb_2$	360	/	0.1 C	5
FeSb-TiC-C	215	210 after 60 cycles	100	6
SnSb nanocrystals	~365	350 after 100 cycles	330	7
SnSb/carbon	347	345 after 205 cycles	100	8
	~420	360 after 100 cycles	100	9
	544	435 after 50 cycles	100	10
Cu-Sb	250	~425 after 100 cycles	0.2 C	11
	616.85	485.6 after 120 cycles	0.1 C	12
Mo_3Sb_7	330	~160 after 100 cycles	3.5 C	13
AlSb	~450	~250 after 50 cycles	1/6 C	14
Sb/C fibers	422	350 after 300 cycles	100	15
	495	446 after 400 cycles	200	16
Sb/carbon microspheres	402	372 after 100 cycles	300	17
Sb/N-carbon nanosheets	340	305 after 60 cycles	50	18
milled Sb/C	610	575 after 100 cycles	100	19
Sb@C structures	230	190 after 10 cycles	10	20
Sb/carbon nanotube	502	382 after 120 cycles	200	21
Sb/graphene	~600	~530 after 150 cycles	328	22
Sb/acetylene black	520	473 after 70 cycles	100	23
Sb/Sb_2O_3/PPy nanowires	466.81 520.64 at 19^{th} cycle	512.01 after 100 cycles	66	24
rod-like Sb/C	630.1	430.9 after 100 cycles	50	25
Pitaya-like Sb@C microspheres	628	~584 after 100 cycles	200	26
Sb_2O_3/Sb@ graphene	527.9	486.5 after 275 cycles	100	27
Sb nanocrystals	~600	~580 after 100 cycles	660	28
Bulk Sb	537	576 after 160 cycles	330	29
Sb hollow nanospheres	627	622.2 after 50 cycles	50	30
Sb porous hollow microspheres	634.6	617 after 100 cycles	100	31
	574.9	502.3 after 120 cycles	660	
CL-Sb	639.2	629 after 120 cycles	100	this work
	556.6	550 after 120 cycles	660	



Scheme S1 Illustration of the formation process of CL-Sb.

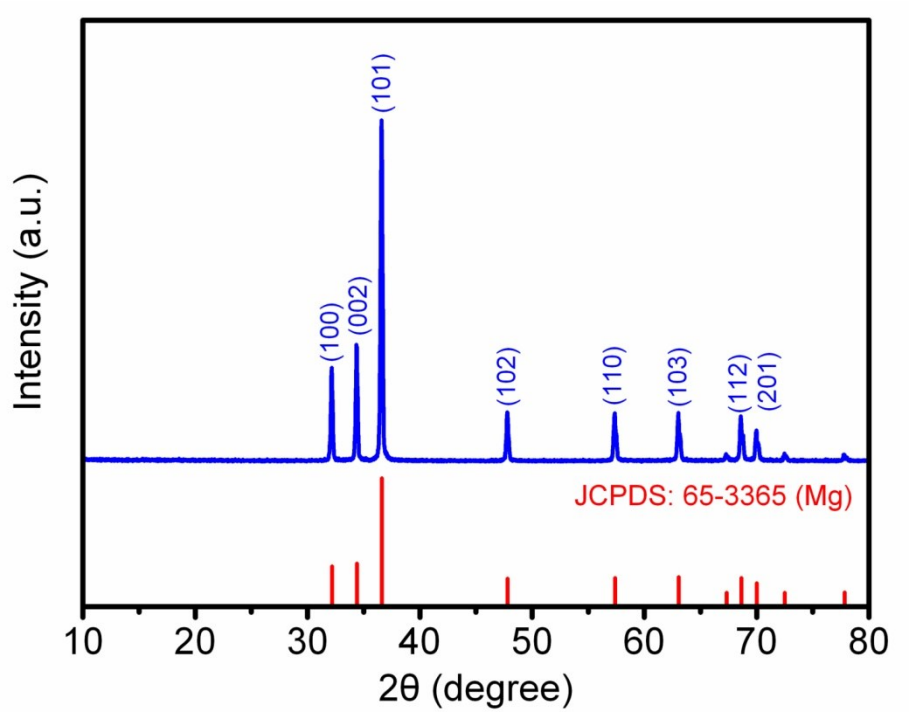


Fig. S1 XRD pattern of Mg powder.

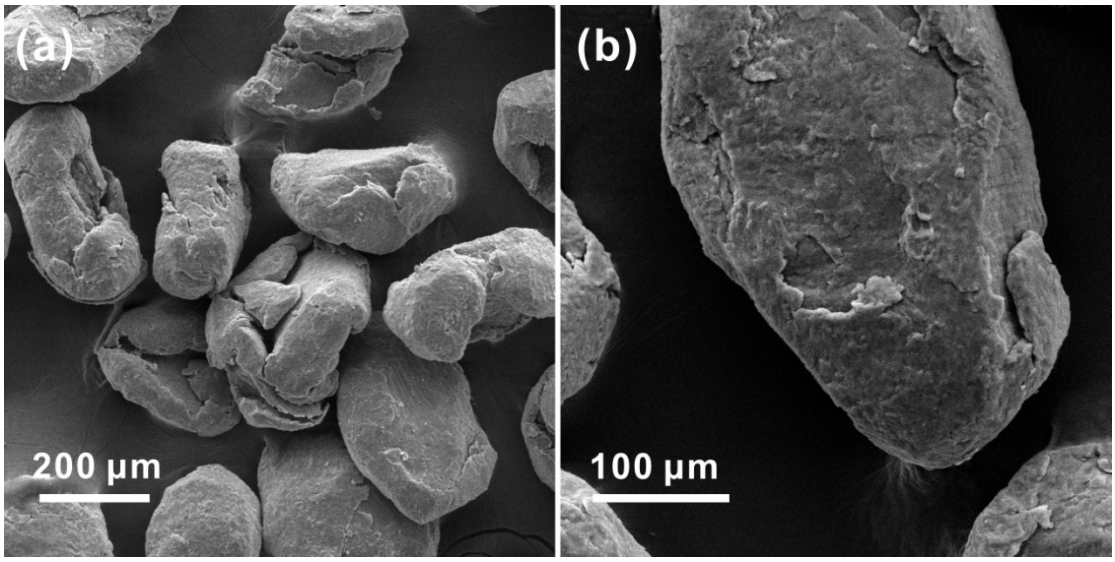


Fig. S2 SEM images of Mg powder.

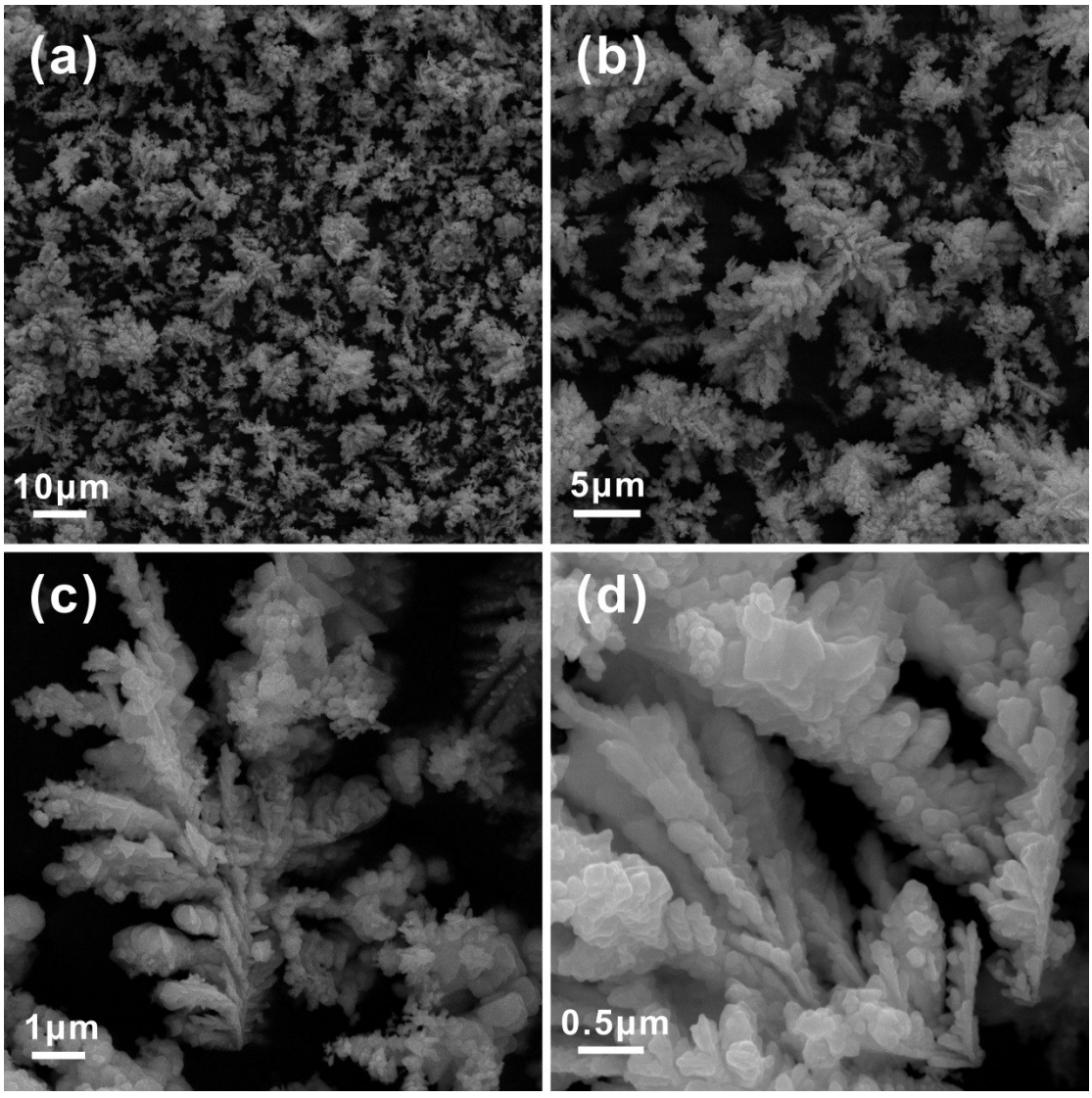


Fig. S3 SEM images of the prepared CL-Sb.

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