

Supporting Information

Super long-life potassium-ion batteries based on antimony@carbon composite anode

Qian Liu,^{ab} Ling Fan,^b Ruifang Ma,^b Suhua Chen,^b Xinzhi Yu,^b Hongguan Yang,^b Yong Xie,^c Xu Han,^{a*} and Bingan Lu^{b*}

Section SI. Experimental Section

Synthesis of Sb@G@C. The detail of synthetic procedure for the Sb@G@C is listed as follows: first, 1g SbCl₃ (Ar., 99.9%) and 8g citric acid (Ar., 99%) was dissolved in deionized water (10 mL) to obtain a transparent homogeneous solution. Then, GO (200 mg, 5mg ml⁻¹) was added into the above solution. After stirring for 30 min, the as-obtained homogeneous solution was rapidly frozen. Subsequently, the material was freeze-drying and the obtained sponge-like sample was ground, and then the obtained material was subjected to heat treatment. Specifically, the composite powder was placed in a quartz boat located in a tube furnace and heated at 550, 650 and 750°C for 2 h with a heating rate of 5 °C min⁻¹ under H₂/Ar (v/v=10/90), respectively. For comparison, the Sb@C was prepared the same as Sb@G@C except for none GO was added, Sb@G was also prepared the same as Sb@G@C except for none citric acid was added, and the heating temperature for Sb@C and Sb@G was 650 °C.

Material characterization. The microstructure morphology of Sb@G@C was studied by TEM (Tecnai F20). The morphology and structure of Sb@G@C was characterized by SEM (Hitachi, S-4800, 5kV). XRD (Philips, X'pert pro) spectrum was conducted with Cu K(alpha) Ka radiation (= 0.154184 nm). We used Ni filters to remove K(beta). The precision is 0.01 mg of the scale (CPA225D Sartorius) for weighing the electrode.

Electrochemical measurements. The cathode was prepared by mixing active materials (Sb@G@C, Sb@C, Sb@G, and pure Sb) with conductive carbon black, and sodium carboxymethylcellulose at a weight ratio of 8:1:1, then paste onto copper foil with mass loading (active material) of 1.50 (±0.20) mg cm⁻², and then dried at 80°C for over 12 h. Organic electrolyte—3M potassium bis(fluoro-sulfonyl)imide (KFSI) in dimethyl ether (DME) was prepared as previously literatures. Half cells of the PIBs were assembled with 2032 coin cells in glovebox under Ar with Sb@G@C as the anode, potassium foil as the counter electrode, Whatman glass fibers were used as the separator and 3M KFSI in DME as the electrolyte. Galvanostatic charge/discharge and rate performance of the battery were evaluated by Arbin Instrument BT 2000 model. CV curves of the batteries were executed by electrochemical workstation. To test the cycled electrodes, the battery was disassembled in the glovebox, followed by washing the electrodes with DME twice to remove the potassium salt.

All the specific capacity of battery was calculated in the base of the composite mass.

In spite of the theoretical capacity of Sb is only 660 mAh g⁻¹, Carbon in the Sb-C composite is also contributing capacity. The capacity of the carbon-based materials usually is around 300 mAh g⁻¹. Therefore, the theoretical capacity of the Sb@G@C composite could be calculated to ~470 mAh g⁻¹.

The Sb@G@C composite was facilely obtained via one-step synthesis strategy. First, the metal salt (SbCl₃) and citric acid was dissolved in deionized water to obtain a transparent homogeneous solution. Then, GO solution was added into the above solution. After stirring for 30 min, the as-obtained homogeneous solution was rapidly frozen. Subsequently, the solid was removed by freeze-drying and the obtained sponge-like sample was ground into fine powder, and then subjected to heat treatment. Specifically, the composite powder was placed in a quartz boat located in a tube furnace and heated at 550, 650 and 750°C for 2 h with a heating rate of 5°C min⁻¹ under H₂/Ar (v/v=10/90), respectively.

Section SII. Supporting Figure

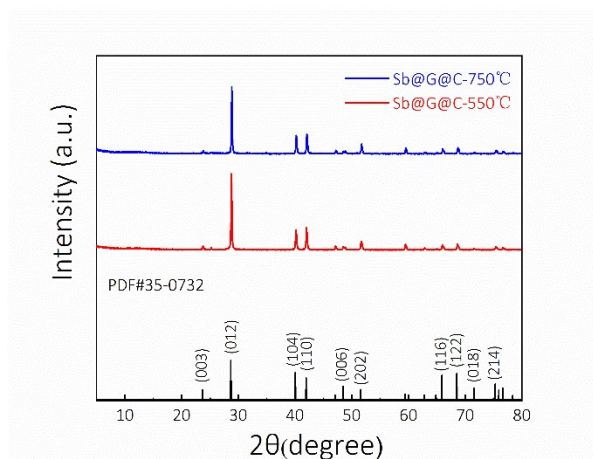


Fig. S1 Powder XRD patterns of Sb@G@C composites at 550 and 750°C.

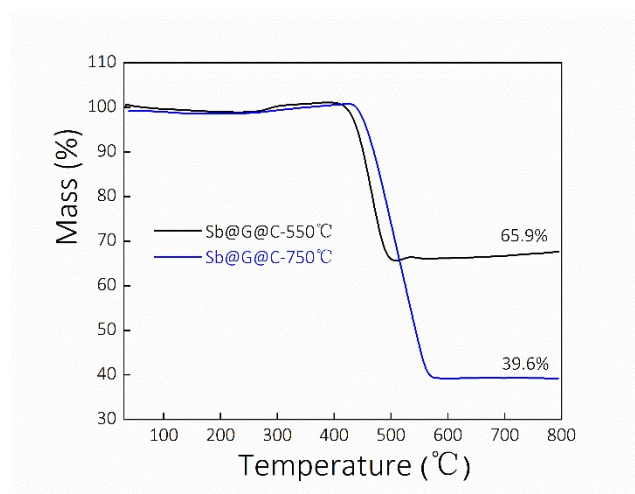


Fig. S2 Thermogravimetric curves (in the atmosphere of flowing air) of Sb@G@C composites at 550 and 750°C.

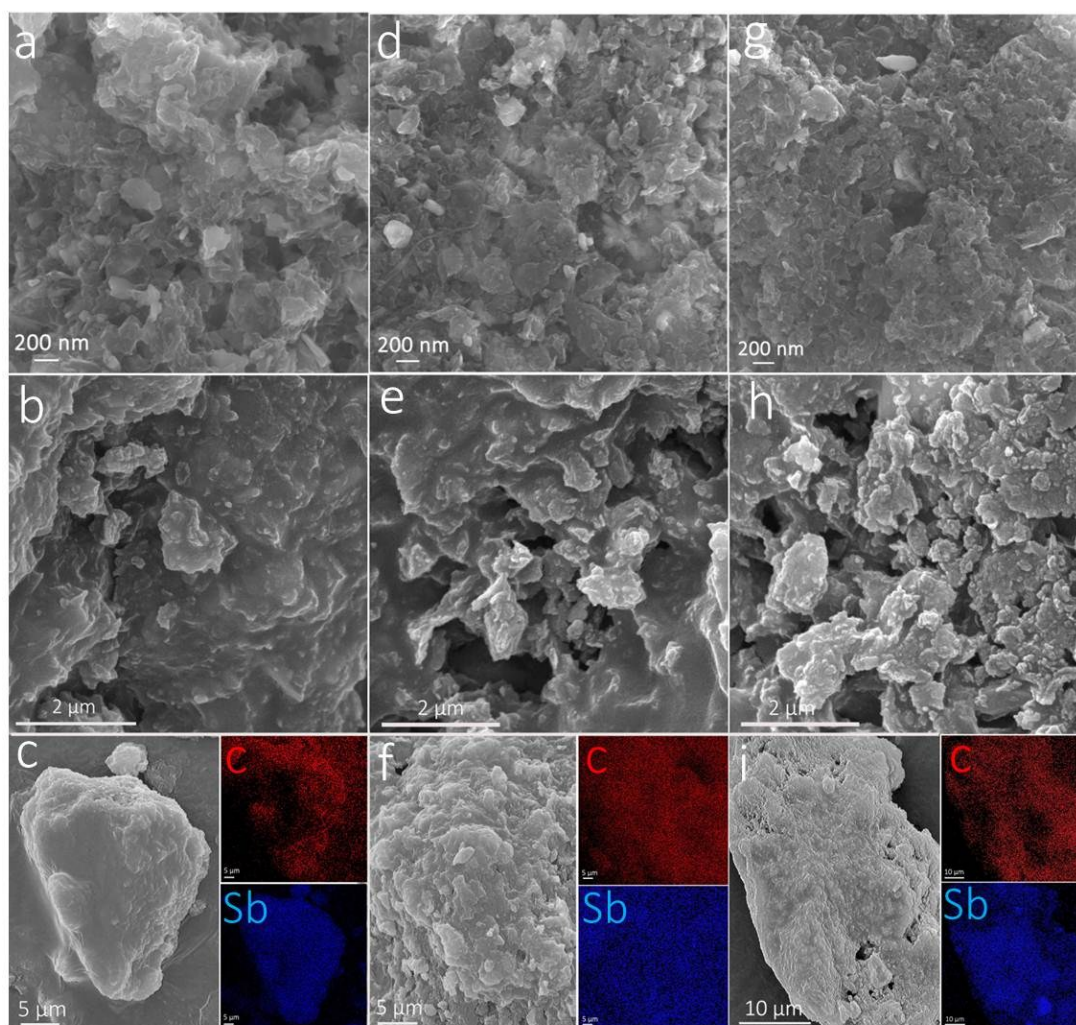


Fig. S3 SEM, low-magnification SEM and EDS elemental mapping images of Sb@G@C composites (a-c) 550°C, (d-f) 650°C, (g-i) 750°C.

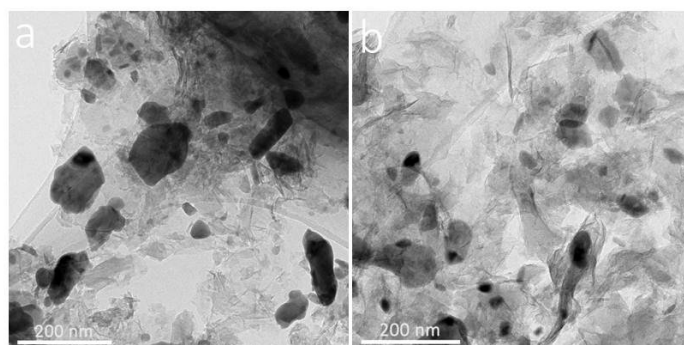


Fig. S4 (a-b) Low magnification TEM image of Sb@G@C composite (550 and 750°C).

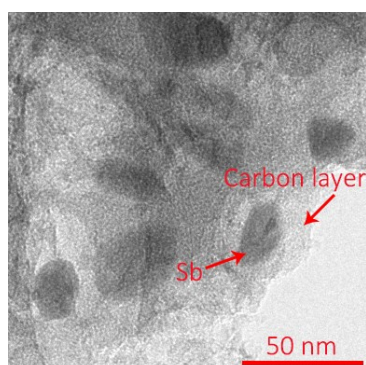


Fig. S5 TEM image of Sb@G@C composite (650°C).

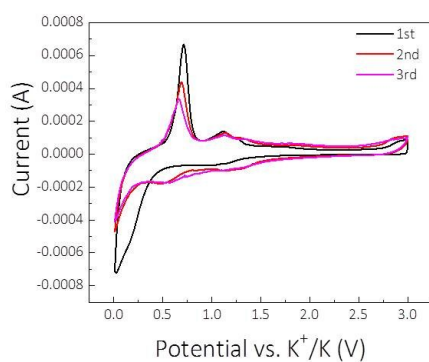


Fig. S6 CV curves of the Sb@G@C (550°C) electrode from 3.0 to 0.01 V at a scan rate of 0.1 mV s⁻¹.

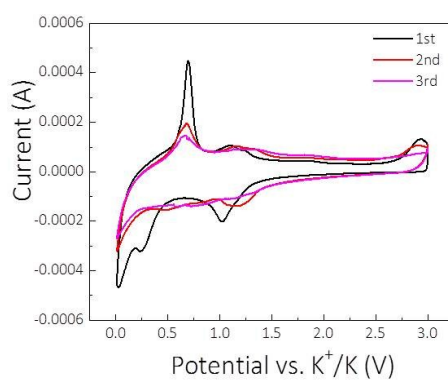


Fig. S7 CV curves of the Sb@G@C (750°C) electrode from 3.0 to 0.01 V at a scan rate of 0.1 mV s⁻¹.

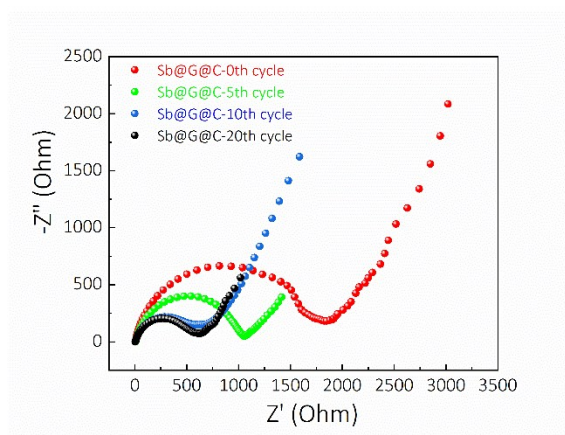


Fig. S8 Nyquist plots of the Sb@G@C (650°C) before cycling after 5, 10 and 20 cycles.

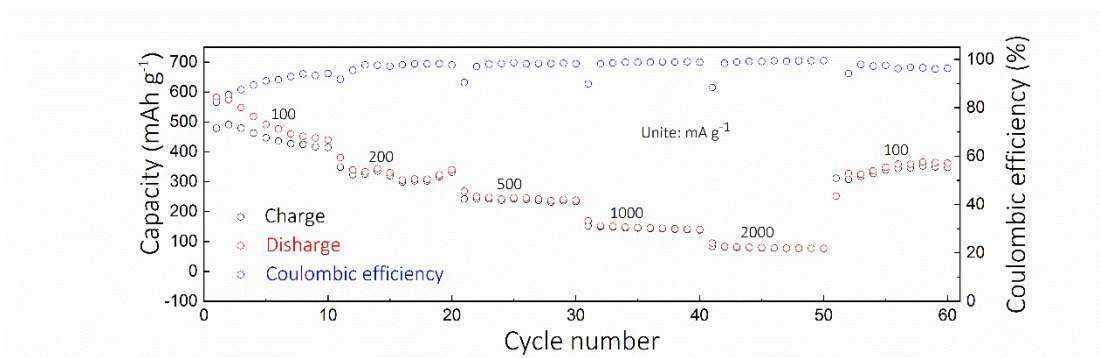


Fig. S9 Rate capacities of Sb@G@C (550°C) electrode at various current rates from 100 to 2000 mA g⁻¹.

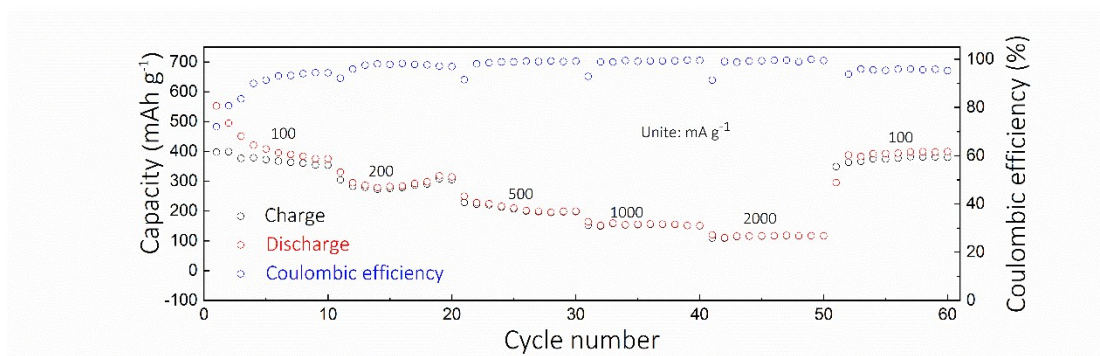


Fig. S10 Rate capacities of Sb@G@C (750°C) electrode at various current rates from 100 to 2000 mA g⁻¹.

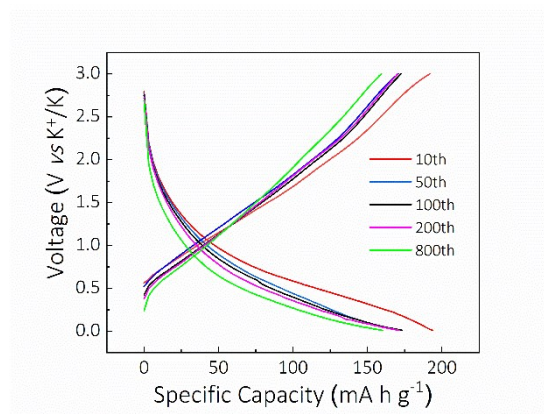


Fig. S11 Charge/discharge profiles of the Sb@G@C (650°C) electrode at a current rate of 1000 mA g⁻¹ between 3.0 and 0.01 V for the 10th, 50th, 100th, 200th, 800th.

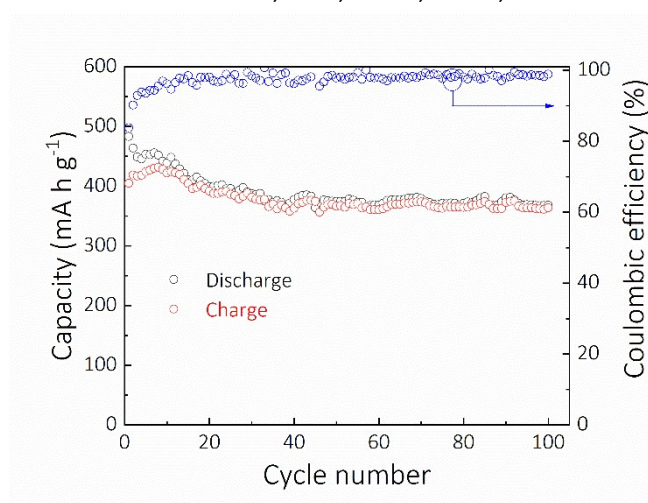


Fig. S12 Cycling performances of Sb@G@C (650°C) electrode at a current rate of 100 mA g⁻¹.

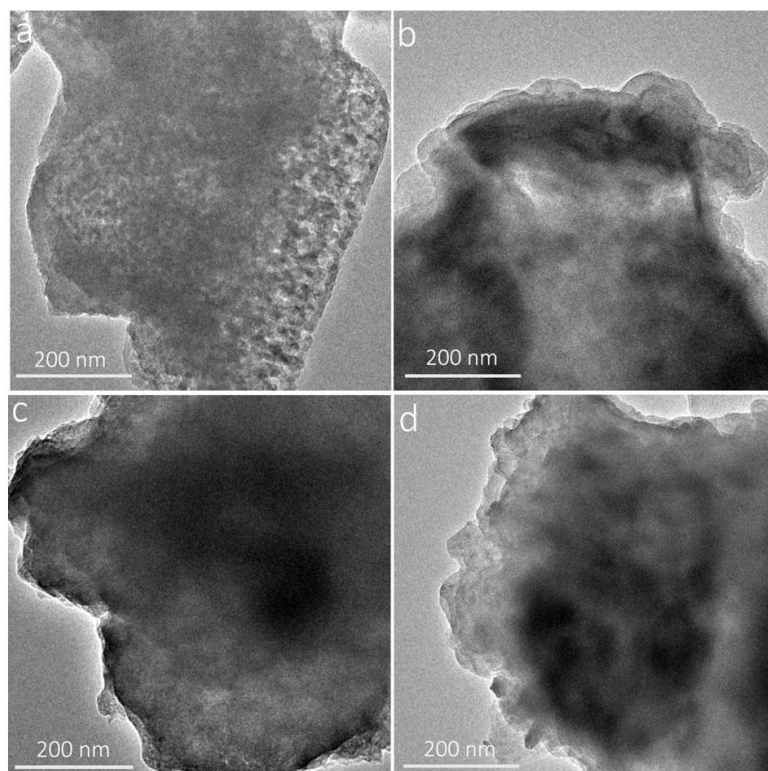


Fig. S13 TEM images of Sb@G@C (650°C) at different states (a) discharged to 0.8 V at the first cycle. (b) discharged to 0.4 V at the first cycle. (c) charged to 0.8 V at the first cycle. (d) charged to 1.4 V at the first cycle.