# Supplementary Information

# A High-Performance Aluminum-Ion Battery with CuS@C Microsphere Composite Cathode

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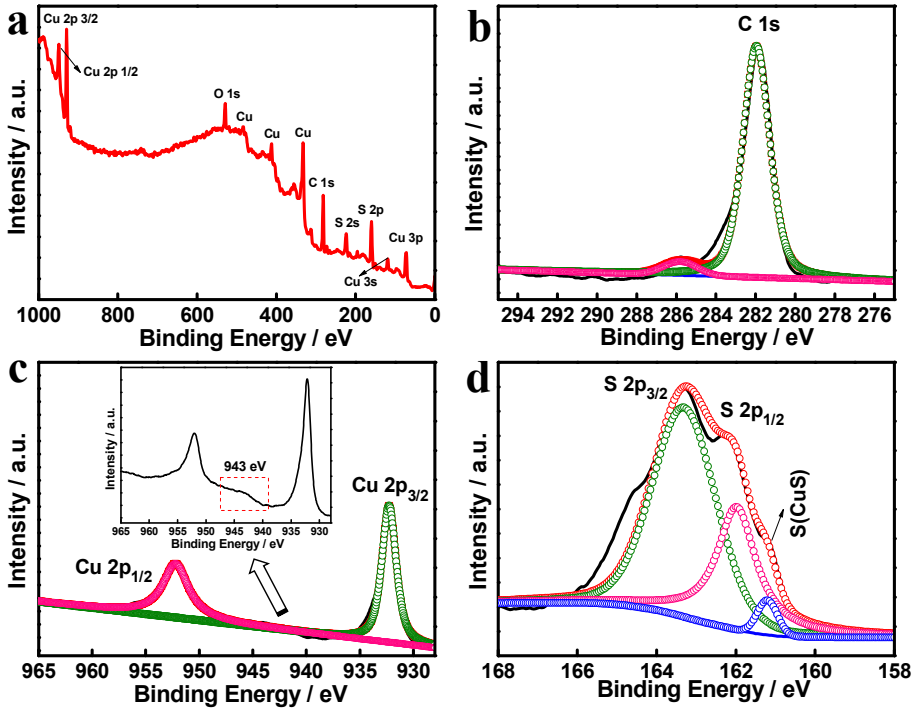


**Figure S1.** Chemical probing of the products by XPS. (a) Wide survey XPS spectrum of the as-prepared CuS microspheres, (b) C 1s spectrum, (c) Cu 2p spectrum, and (d) S 2p spectrum.

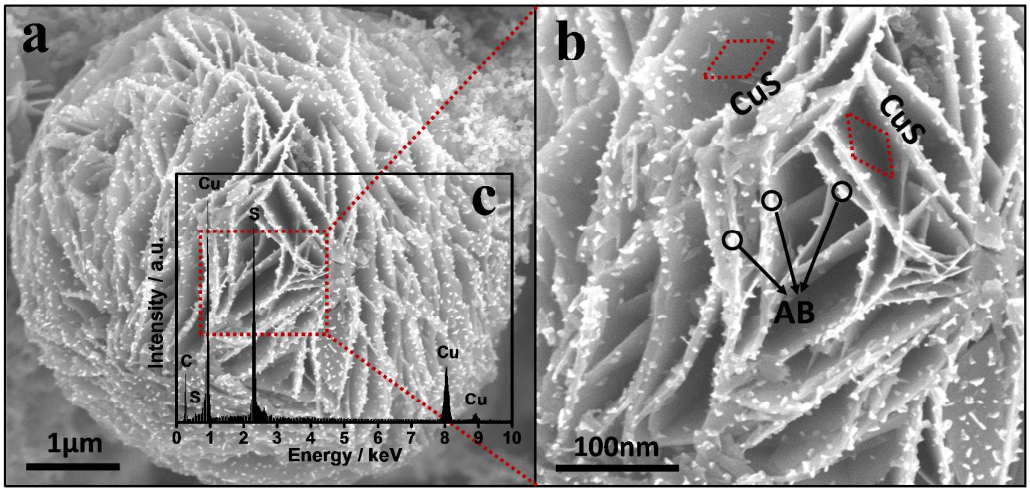


**Figure S2.** Morphologies and compositions of CuS@C microsphere composites. (a,b) FE-SEM images, and (c) The representative EDS spectrum.

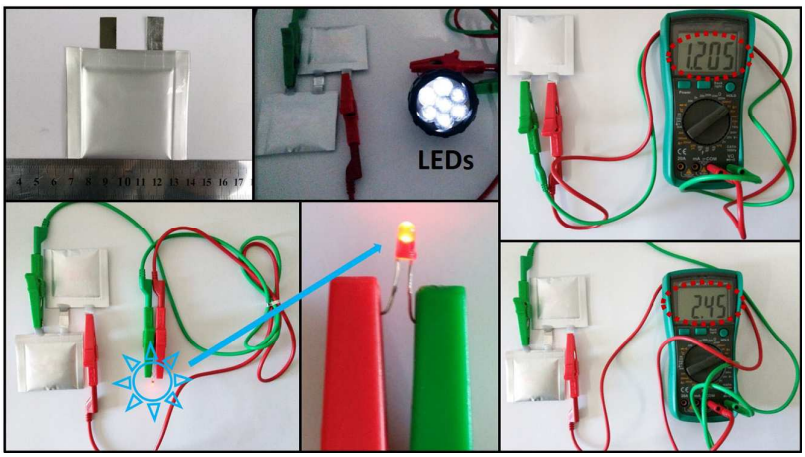


**Figure S3.** The schematic diagram of the soft package aluminum-ion battery and the voltage measurements of the aluminum-ion batteries. And the batteries can light the red LED lamps.

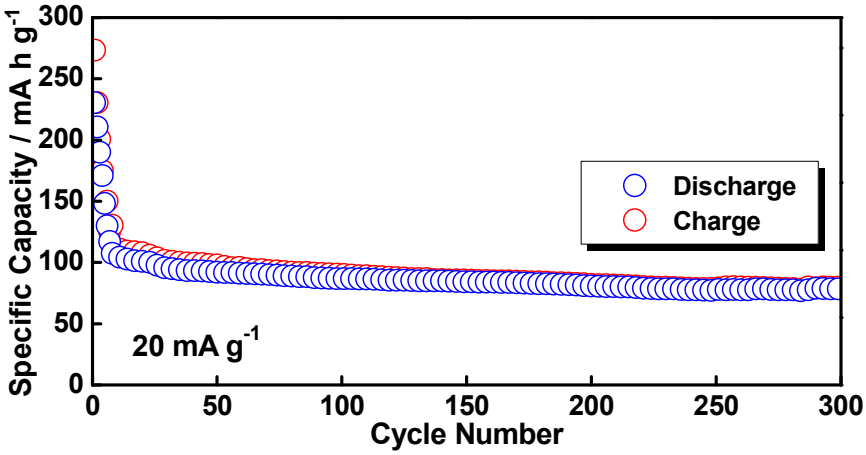


**Figure S4.** Cycling performance of CuS microsphere cathode at a current density of 20 mA $g^{-1}$ over 300 cycles.

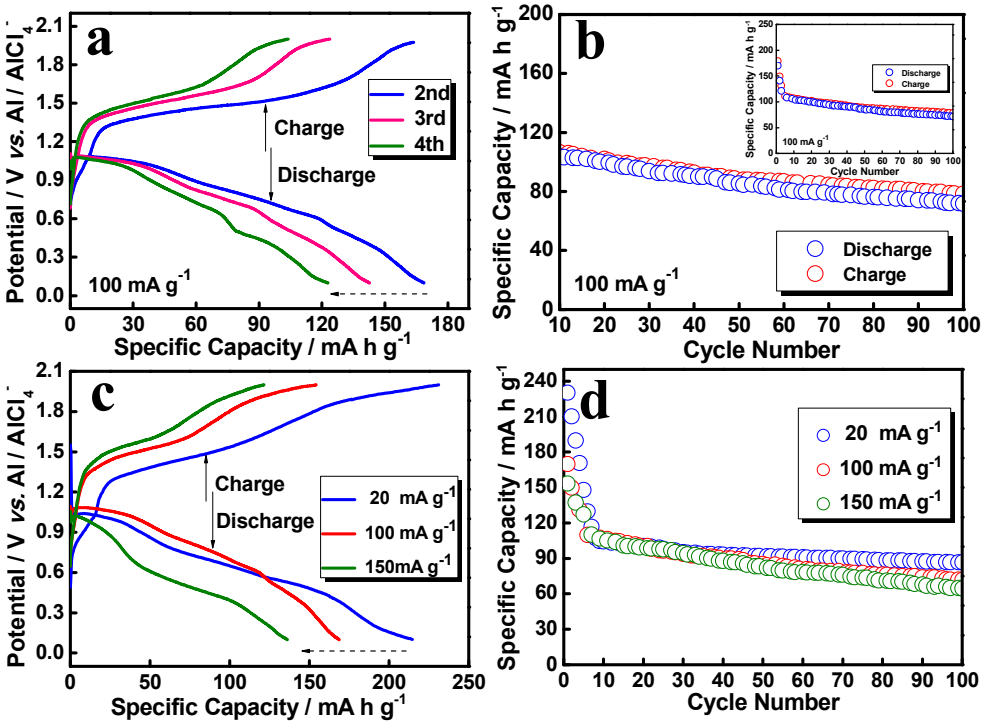


**Figure S5.** Discharge-charge curves and cycling performance of CuS microsphere cathode at different current densities. (a) The typical discharge/charge curves in the voltage range of 0.1~2.0 V *vs*. Al/$AlCl_4^-$ from the 2nd to 4th cycle. (b) The cycling performance at a current density of 100 mA $g^{-1}$ (inset: the cycle number from 0 to 100 cycles). (c) The second discharge/charge curves at different current densities. (d) The cycling performance at various current densities over 100 cycles.

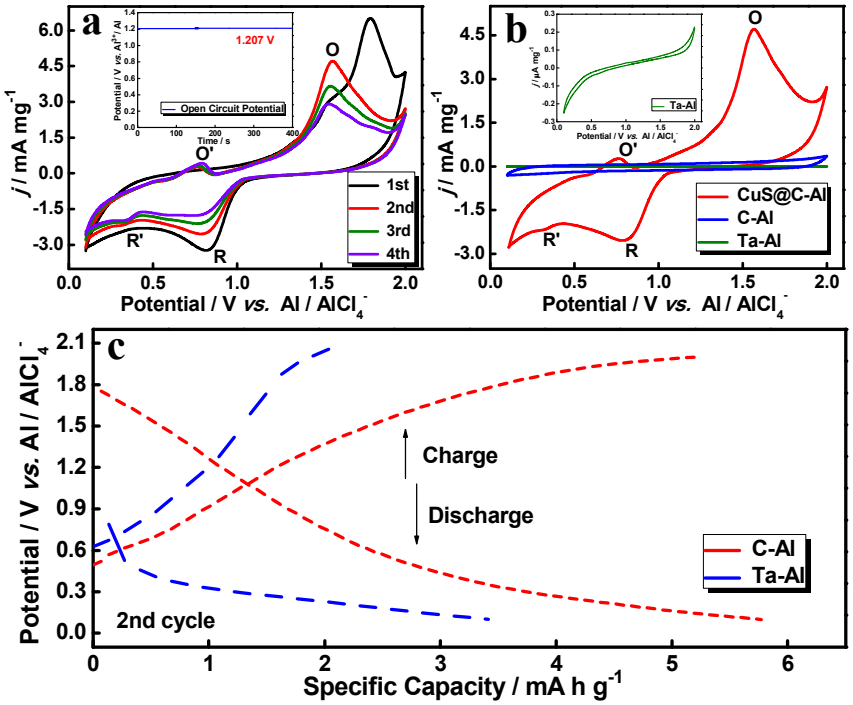


**Figure S6.** (a) Cyclic voltammograms of CuS microsphere cathode at a scan rate of 5 mV $s^{-1}$ between 0.1 and 2.0 V *vs*. $Al/AlCl_4^-$ (inset: open circuit potential). (b) CVs of CuS microsphere electrode, C electrode and Ta electrode, respectively. (c) The second discharge/charge curves of C electrode and Ta electrode in the voltage range of 0.1~2.0 V *vs*. $Al/AlCl_4^-$.

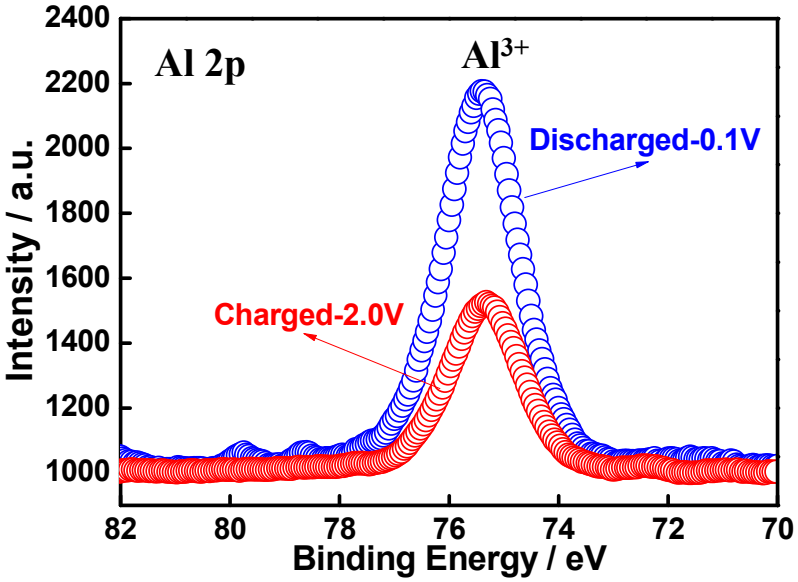


**Figure S7.** Aluminum content analysis of the electrode after cycling. XPS spectra of Al 2p after discharge and charge process.

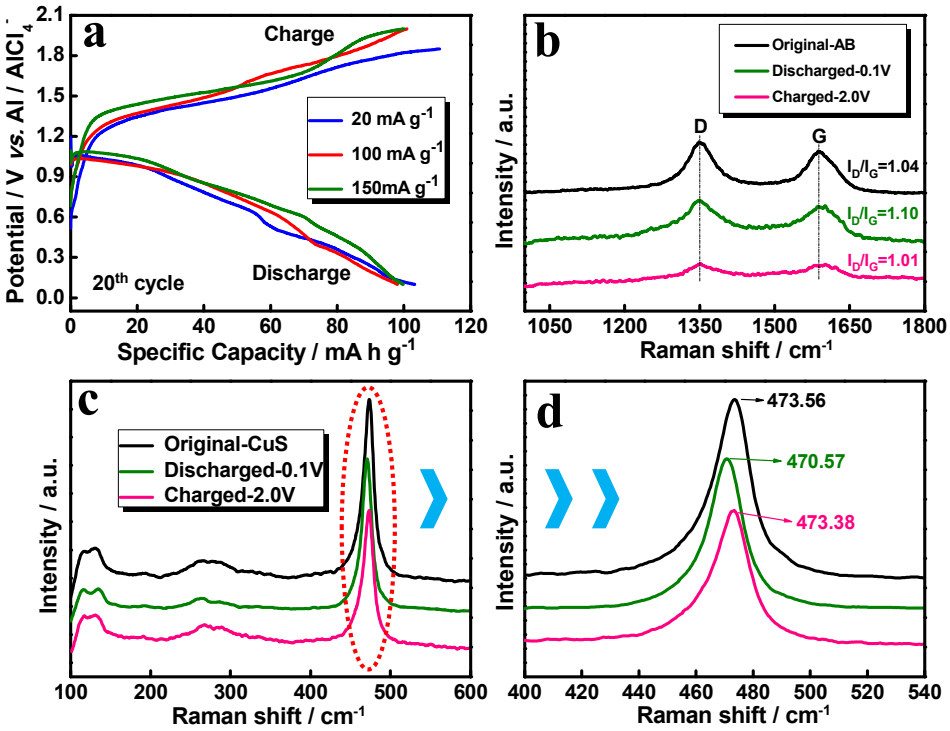


**Figure S8.** (a) The 20th discharge/charge curves in the voltage range of 0.1~2.0 V *vs.* $Al/AlCl_4^-$ at different current densities. (b) Raman spectra of acetylene black before and through the 20th cycle. (c,d) Raman spectra of active materials (CuS) before and through the 20th cycle.

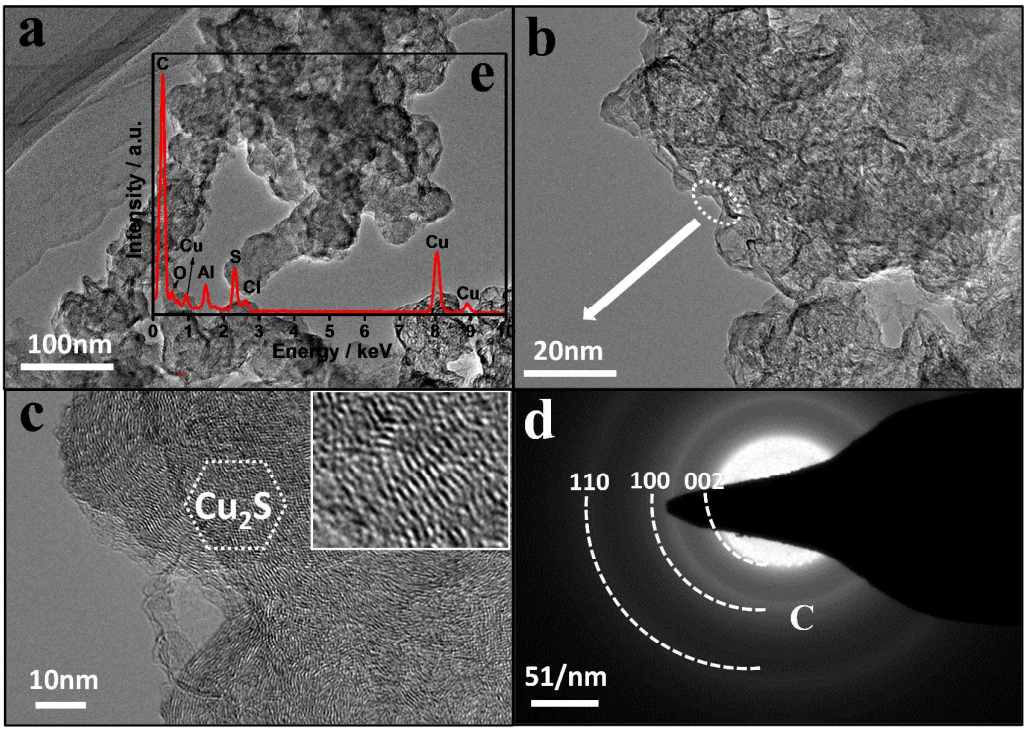


**Figure S9.** Morphologies and compositions of the electrode after discharge process. (a,b) TEM images, (c) HRTEM images, (d) SAED pattern of acetylene black, and (e) The representative EDS spectrum.

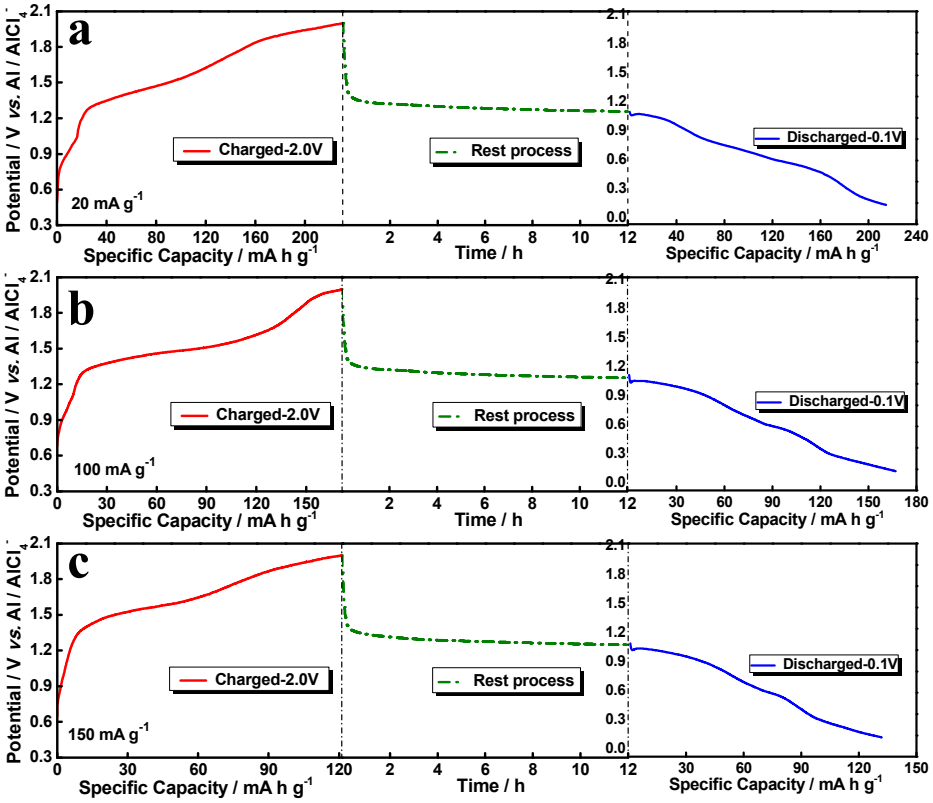


**Figure S10.** Self-discharge behavior of the aluminum-ion battery. The battery was rested for 12 h after charging to 2.0 V *vs*. Al/$AlCl_4^-$ at different current densities. (a) 20 mA $g^{-1}$, (b) 100 mA $g^{-1}$, (c) 150 mA $g^{-1}$.

**Table S1.** Element content of the cathode. EDS analyses results of CuS@C microsphere composites.

| ***Element*** | ***wt. %*** | ***at. %*** |
|---|---|---|
| ***C*** | 29.43 | 62.25 |
| ***Cu*** | 46.29 | 18.49 |
| ***S*** | 24.28 | 19.26 |
| ***Total*** | 100 | 100 |