

SUPPORTING INFORMATION

Single-Crystalline and Hierarchical VSe₂ as Aluminum-ion Battery Cathode

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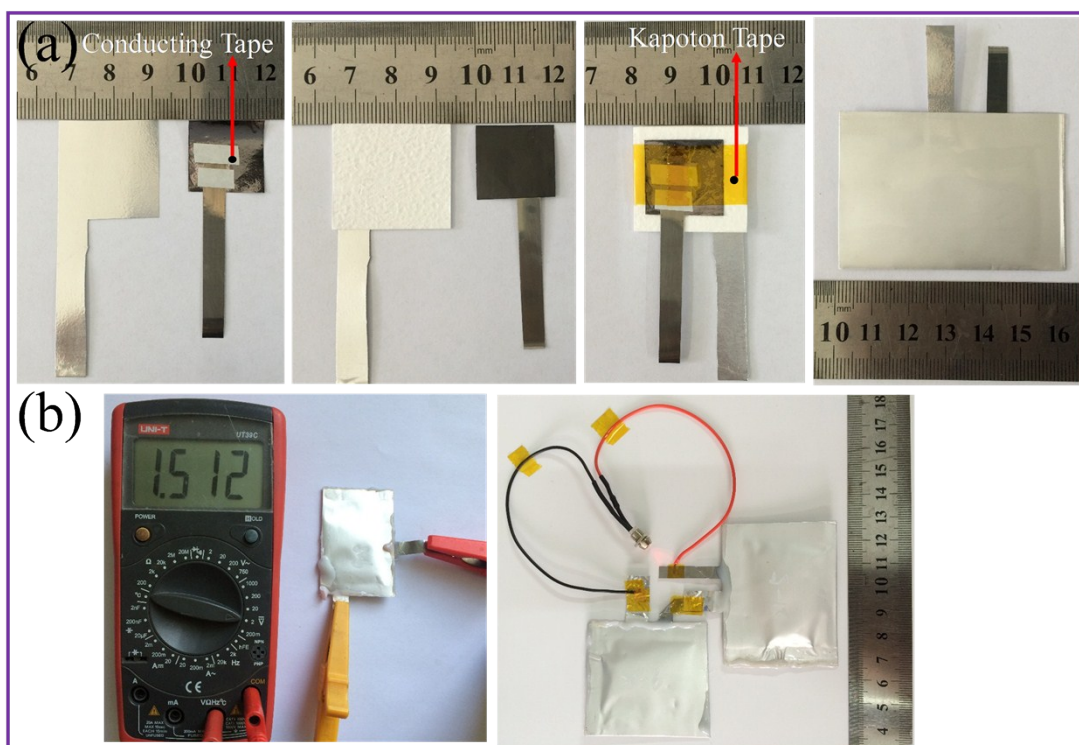


Fig. S1 The assembly schematic diagram of soft package aluminum-ion battery.

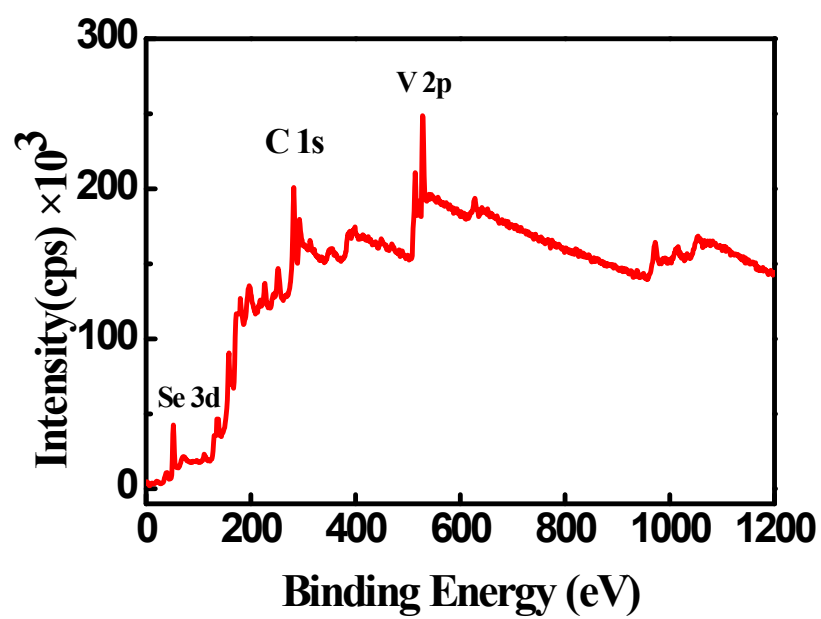


Fig. S2 The wide survey XPS spectra of as-prepared VSe₂.

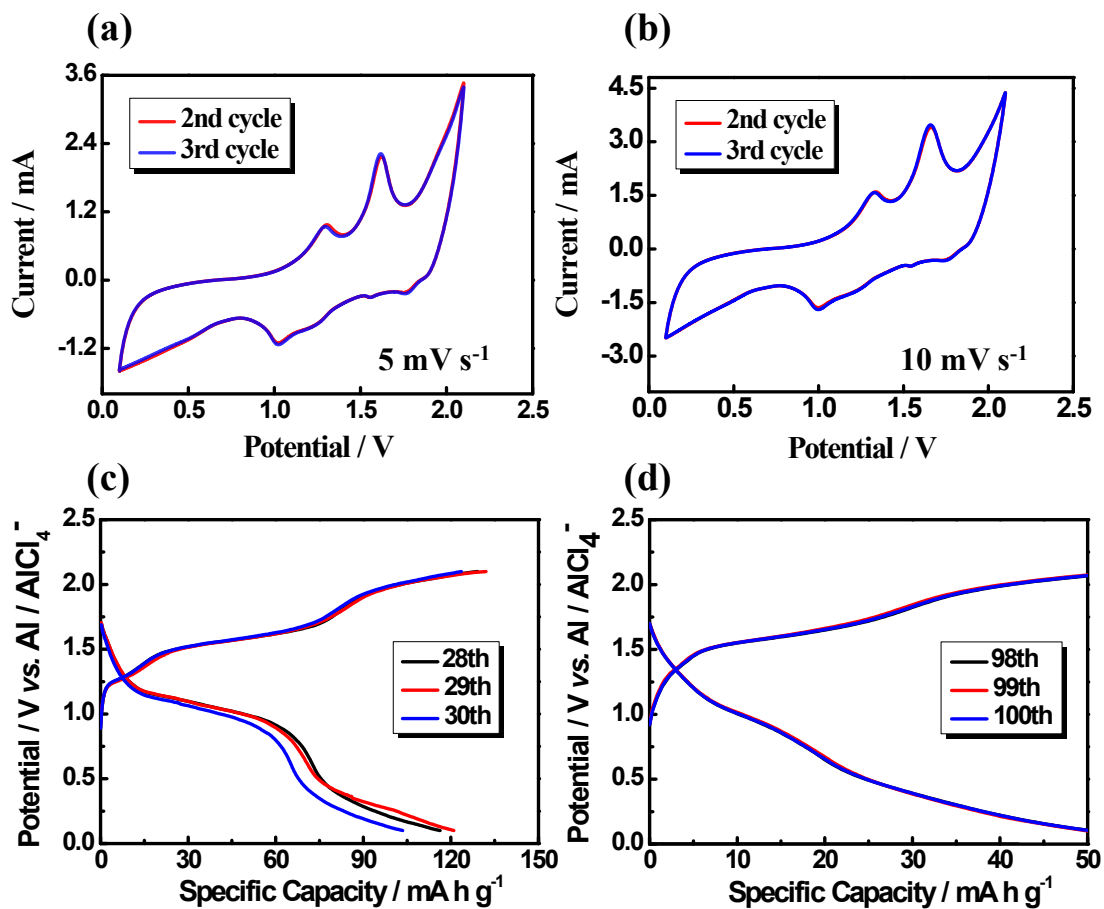


Fig. S3 (a) CV curves for the initial three cycles at a scan rate of 5 mV s⁻¹. (b) CV curves for the initial three cycles at a scan rate of 10 mV s⁻¹. (c) The charge/discharge curves for the 28th-30th cycles at a current density of 100 mA g⁻¹. (d) The charge/discharge curves for the 98th-100th cycles at a current density of 100 mA g⁻¹.

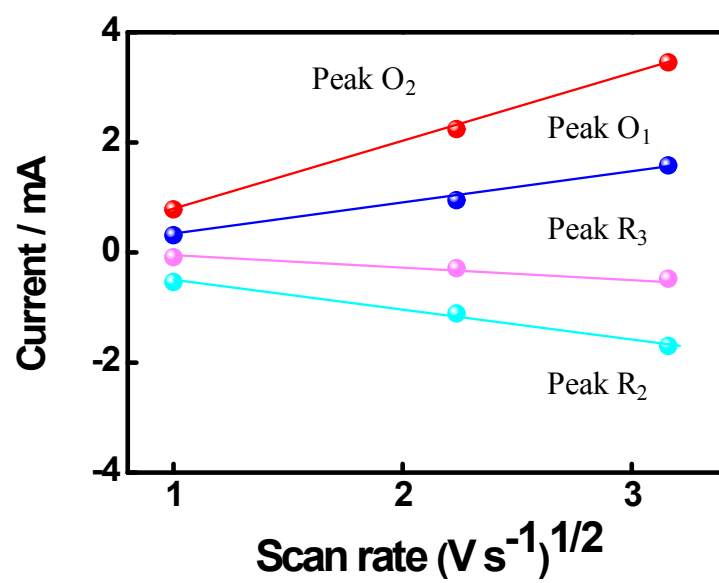


Fig. S4 Relationships between the current peak values and the scan rates (I_p vs. $V^{1/2}$).

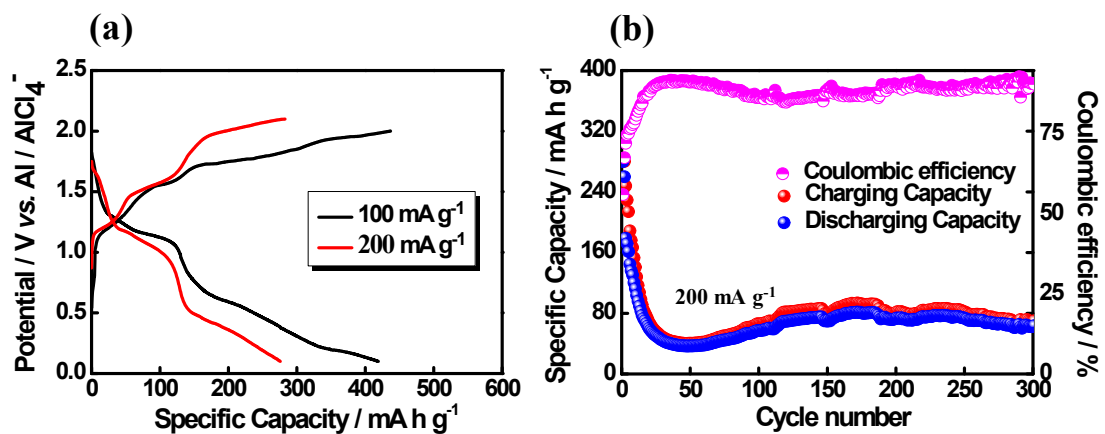


Fig. S5 (a) The second charge/discharge curves at 100, 200 mA g⁻¹ current densities. (b) The cycling performance at a current density of 200 mA g⁻¹.

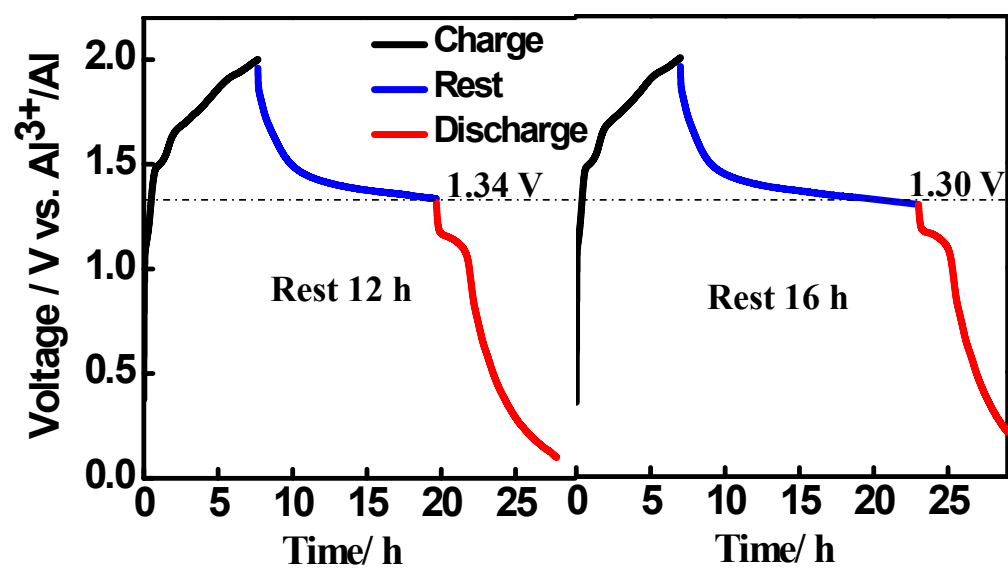


Fig. S6 The self-discharge curves of the aluminum-ion battery by resting 12 and 16 h.

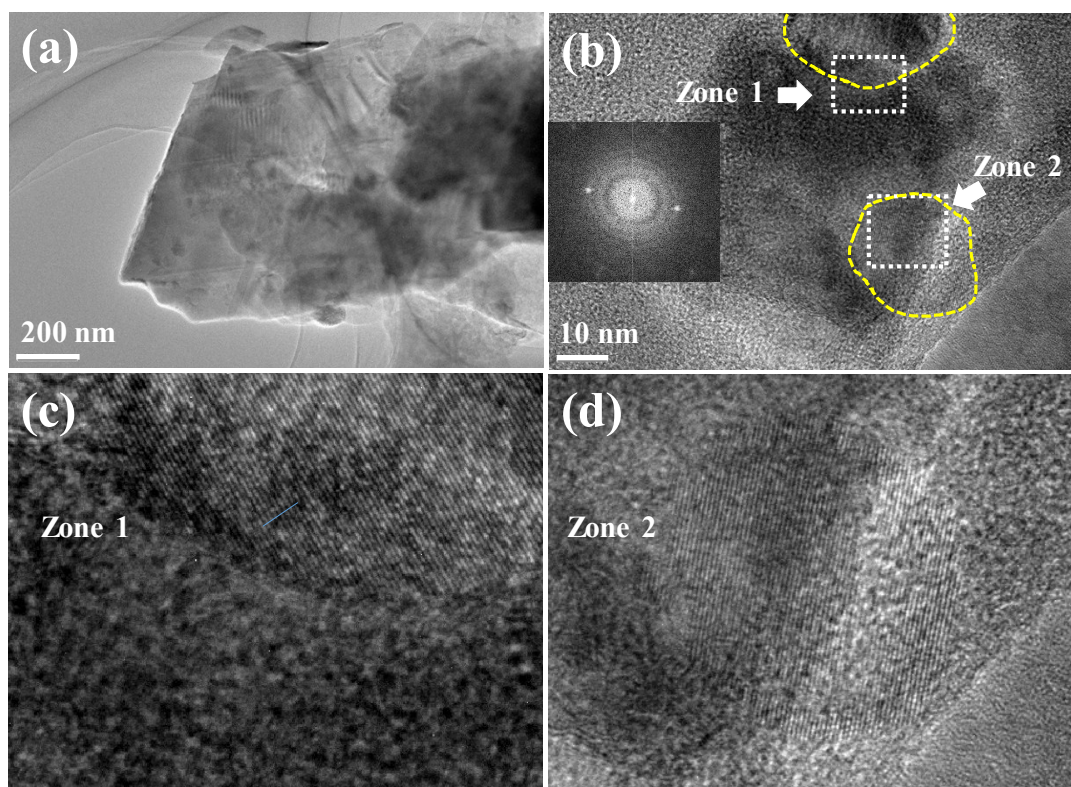


Fig. S7 (a) TEM and (b) HRTEM image of the cycled cathode. Inset: Fast Fourier transform of the image. (c) The enlargement of zone 1 in (b). (d) The enlargement of zone 2 in (b).

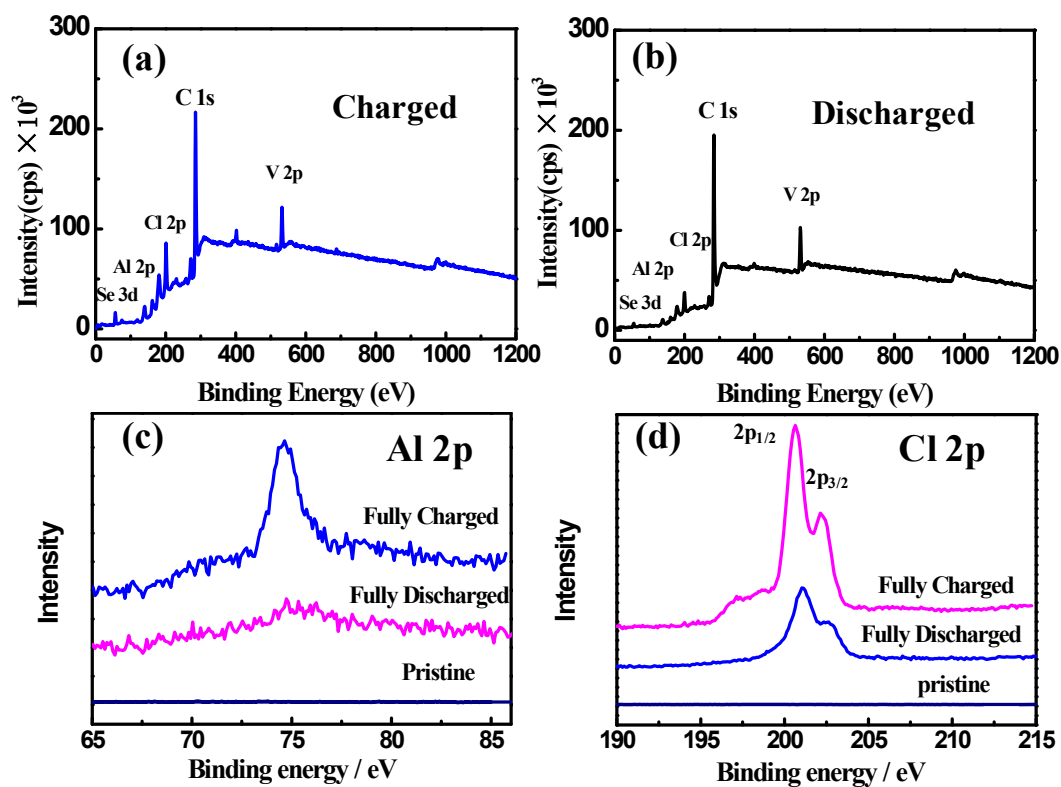


Fig. S8 The wide survey XPS spectra of the (a) charged and (b) discharged cathode. (c) Al 2p and (d) Cl 2p peaks of the cathode: pristine, fully charged and fully discharged.

Table S1. EIS analyses results of the battery based on the VSe₂ cathode at the zeroth, 10th charged, and 10th discharged, respectively.

Samples	Rs (Ω)	Rct (Ω)	CPE ₁ - T($\times 10^{-5}$)	CPE ₁ -P	Rp(Ω)	Wo-R	Wo-T($\times 10^{-3}$)	Wo-P
Fresh cell	9.40	7.66	9.08	0.81	10.01	3.04	1.71	0.41
Charged	12.11	12.52	8.55	0.80	16.90	6.42	3.84	0.39
Discharged	10.91	9.80	15.78	0.76	15.10	6.86	4.15	0.40