Supporting Information

Long-cycle and high-rate electrochemical performance of expanded graphite cathode materials with two-stage aluminum storage

mechanism

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Fig.S1. (a) The nitrogen adsorption/desorption isotherms and pore-size distributions of the P-EG; (b-d) the pore-size distributions of the 700- EG, 800-EG and 900-EG.



Fig. S2. (a, b) SEM micrographs of the P-EG with different magnifications; (c, d) SEM micrographs of the 700-EG with different magnifications; (e, f) SEM micrographs of the 900-EG with different magnifications.



Fig. S3. (a-c) TEM image in the 700-EG; (d-f) TEM image of the 900-EG.



Fig. S4. SEM micrographs of the 800-EG electrode after 10 cycles (a) and after 10000 cycles (b).



Fig. S5. *In-situ* XRD data of the graphite electrode in AGBs under charging and discharging to various voltages under 0.5 A g^{-1} for the third cycle.



Fig. S6. (a) The CV curve of the 800-EG electrode in the voltage range of 0.5-1.5 V; (b) the charge-discharge curves of the 800-EG electrode at 0.5 A g^{-1} at 0.5-1.5 V; (c) the CV curve of the 800-EG electrode in the voltage range of 1.5-2.5 V; (d) the charge-discharge curves of the 800-EG electrode at 0.5 A g^{-1} at 1.5-2.5 V.



Fig. S7. (a) corresponding log (i) versus log (v) plots at specific peak currents; (b) CV curve with the capacitive contribution fraction shown by the shaded area at a scan rate of 5 mV s⁻¹ scan rates of the 800-EG at 0.5-2.5.



Fig. S8. (a, b) corresponding log (i) versus log (v) plots at specific peak currents of 700-EG and 900-EG electrode (1.5-2.5V); (c, d) bar chart showing the percent of capacitive contribution at different scan rates of 700-EG and 900-EG electrode.

Calculation Process:

The diffusion coefficient of $AlCl_4^-$ ($D_{AlCl_4^-}$) in the expanded graphite electrode material can be obtained by the following formula:

$$Zre = Rct + Rs + \sigma \omega^{-1/2}$$
(1)
$$D_{AlCl4^{-}} = \frac{(RT)^{2}}{2A^{2}n^{4}F^{4}C_{AlCl4^{-}}^{2}\sigma^{2}}$$
(2)

Where R is the gas constant (8.314J K⁻¹ mol⁻¹); T is the absolute temperature (293.15 K, 313.15K and 333.15K); A is the surface area of the positive (1 cm²), n is charge transfer number of AlCl₄⁻; F is the Faraday constant (96000 C mol⁻¹); $AlCl_4^-$ is the concentration of AlCl₄⁻ in the electrode, ω is the angular frequency, and σ is the Warburg factor, which is relative to Z_{re}. The value can be obtained from the slope of the lines in **Figure 5**.