## **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  $0.5 \text{ A g}^{-1}$  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<sup>-1</sup> 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)  
$$D_{AlCl4^{-}} = \frac{(RT)^2}{2A^2n^4F^4C_{AlCl4^{-}}^2\sigma^2}$$
(2)

Where R is the gas constant (8.314J K<sup>-1</sup> mol<sup>-1</sup>); T is the absolute temperature (293.15 K, 313.15K and 333.15K); A is the surface area of the positive (1 cm<sup>2</sup>), n is charge transfer number of AlCl<sub>4</sub><sup>-</sup>; F is the Faraday constant (96000 C mol<sup>-1</sup>);  $AlCl_4^-$  is the concentration of AlCl<sub>4</sub><sup>-</sup> in the electrode,  $\omega$  is the angular frequency, and  $\sigma$  is the Warburg factor, which is relative to Z<sub>re</sub>. The value can be obtained from the slope of the lines in **Figure 5**.