## **Supporting Information**

## A long-life rechargeable Al ion battery based on molten salts

Yang Song<sup>1</sup>, Shuqiang Jiao<sup>1\*</sup>, Jiguo Tu<sup>1</sup>, Junxiang Wang<sup>1</sup>, Yingjun Liu<sup>2\*</sup>, Handong Jiao<sup>1</sup>, Xuhui

Mao<sup>3\*</sup>, Zhancheng Guo<sup>1</sup> and Derek J Fray<sup>2</sup>

<sup>1</sup>State Key Laboratory of Advanced Metallurgy, University of Science and Technology Beijing,

Beijing 100083, PR China. \*Corresponding author: sjiao@ustb.edu.cn (S Jiao)

<sup>2</sup>Department of Materials Science and Metallurgy, University of Cambridge, 27 Charles Babbage Road,

Cambridge, CB3 0FS, UK.\*Corresponding author: yjl26@cam.ac.uk (Y Liu)

<sup>3</sup>School of Resources and Environmental Science, Wuhan University, Wuhan 430072, PR China.

\*Corresponding author: clab@whu,.edu.cn (X Mao)



**Fig.S1(a)** The photograph of a assembled Al/graphite cell;**(b)** The photographs of the aluminum foil (upper), and the graphitic carbon cathode (bottom): original graphitic carbon paper (left), wrapped by glass fiber (middle), the one after10000 cycles (right).



Fig.S2The XRD(a) pattern and SEM image(b) of the pristine graphitic carbon paper before test



Fig.S3 Phase equilibrium diagram of AlCl3-NaCl mixture.<sup>1-3</sup>



**Fig.** S4(a)X-ray diffraction pattern of the inorganic molten salt electrolyte with a mole ratio of  $AlCl_3/NaCl \sim 1.63$ . (b)Raman spectrum of the inorganic molten salt electrolyte with a mole ratio of  $AlCl_3/NaCl \sim 1.63$ .



Fig.S5Theoretical decomposition potentials of theindicated reactions at different temperatures.



**Fig.S6**Coulombic efficiency and capacity retention of an Al/Graphite battery at various current  $densities(100 \sim 500 m Ag^{-1})$ .



**Fig. S7 (a)** The discharge capacity and the corresponding retention rate of the Al/Graphite battery; (b)Galvanostatic charge and discharge curves of Al/Graphite battery charging at 4000mAg<sup>-1</sup> and discharging at various current densities ranging from 100 to 4000 mAg<sup>-1</sup>.(c)The cycling performance and coulombic efficiency at a charging current density of 3000mAg<sup>-1</sup> and a discharging current density of 200mAg<sup>-1</sup>



Fig, S8Surface analysis of the Al anodes, (a-c)SEM images of the Al anodes obtained from threeAl/Graphite battery after 20(a), 100 (b) 10000 (c) cycles, respectively, indicating no dendrite formation over these cycles.



Fig. S9 The discharge capacities of the cells with different aluminum anodes (from 0.0304 to 0.243g) at a current density of 500 mAg<sup>-1</sup>



Fig. S10 SEM image (A) and (B) TEM image of the graphitic carbon papersafter 10000 cycles.



**Fig.S11** (a) The charge–discharge curves at  $100mAg^{-1}$  with a electrolyte of molten salts at  $120 \ C$ ; (b) The charge–discharge curves at  $100mAg^{-1}$  with a electrolyte of ionic liquid at  $120 \ C$ ; (c) Specific capacity of different electrolyte at different current density, when the temperature is  $120 \ C$ 



Fig. S12The cyclic voltammogram curve of an Al-ion batteryat a scan rate of 10 mVs<sup>-1</sup>



*Fig. S13 (a)* Polarization curve of the molybdenum sheet in NaAlCl<sub>4</sub> electrolyte; (b)Charge–discharge curves of Mosheet at 500mAg-1. Theinset shows the charge and discharge cycles.

Table ST   The voluge diop (AD) under different die eut off voluges			
Charge cut-off voltage/V	1.80	1.98	2.14
The voltage drop, $\Delta E/V$	0.352	0.081	0.051

**Table S1** | The voltage drop ( $\Delta E$ ) under different the cut-off voltages



**Fig. S14.(a)** The Nyquist plots of the Al-ion full battery at different bias from 0.0 V to 1.0 V. (b) The linear fits of the Z' vs.  $\omega^{1/2}$  in the low-frequency region. (c) The Nyquist plots of the Al-ion full battery at the 10000th cycle

## Reference

- 1 J. Kendall, E. D. Crittenden and H. K. Miller, J. Am. Chem. Soc., 1923, 45, 963.
- 2 R. Midorikawa, J. Electrochem. Soc. Japan., 1955, 23, 72.
- 3 E. Dewing, Metall. Mater. Trans. B, 1981, 12, 705.