

Supporting Information

Sub-100 °C aluminum ion battery based on ternary inorganic molten salt

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Experimental

Materials

The AlCl₃/LiCl/KCl molten salt electrolyte in this study was prepared by mixing AlCl₃ (99%, Macklin), LiCl (99.99%, Aladdin) and KCl (99.9%, Aladdin) under a molar ratio of 59/29/12 inside an argon glovebox (O₂ and H₂O contents < 0.1 ppm). Anhydrous AlCl₃ and LiCl were used as received, while KCl was pre-dried in a vacuum oven at 80 °C overnight. Aluminum foil (99.9%, 0.05 mm, Beijing Trillion Metals, China) and molybdenum foil (99.9%, 0.05 mm, Beijing Trillion Metals, China) were used as anode and current collector, respectively, which were ultrasonically cleaned by anhydrous ethanol for three times and then dried in a vacuum oven at 80 °C overnight before use. Al foam anode (99.9%, TaiLi Porous Metal, China) was used after washing by anhydrous ethanol with no ultrasonication. Graphite paper cathode (Hua Tai Graphite, China) and glass fiber membrane separator (GF/A, Whatman) were dried in vacuum oven with no washing.

Assembly of batteries

AIBs were assembled inside an argon glovebox. Before moving to the glovebox, all the setups were dried in an oven at 55 °C. A home-made pyrex glass jar (100 mL) coupled with a Teflon cap and well-matched electrode cables was used as the cell (Figure 1b), which was added with a uniform mixture of AlCl₃, LiCl, and KCl (total weight: 40 g) as well as a stirrer bar. Graphite paper was cut into a small piece (1×1 cm², ~8 mg) as the cathode, which was well covered with a piece of separator (1×1

cm²). The cathode and separator were then placed onto a Mo foil piece (1×1.3 cm²) and fastened by folding the Mo edge. The Al anode was prepared by tightly fastening a piece of Al foil (1.5×1.5 cm²) or foam onto a platinum plate. The batteries were well assembled and sealed, and then moved outside the glovebox. The batteries were then heated inside a fume hood under stirring in an oil bath to target temperatures. After a liquid ternary electrolyte was obtained, the battery was maintained for several hours for activation prior to the electrochemical measurements.

Characterization

Electrochemical measurements of AIBs were conducted using a battery testing instrument (Wuhan Land Electronics, China) at various temperatures. The temperature of the oil bath was calibrated prior to use with an error of no more than 2 °C. The specific capacity and current density were calculated based on the mass of the graphite paper. Cyclic voltammogram (CV) curves were acquired using a Solartron electrochemical workstation. Ex-situ Raman spectroscopy was performed on a Renishaw Raman analyzer, with a laser wavelength of 532 nm. Scanning electron microscopy (SEM) was conducted using a Hitachi S-4800 instrument.

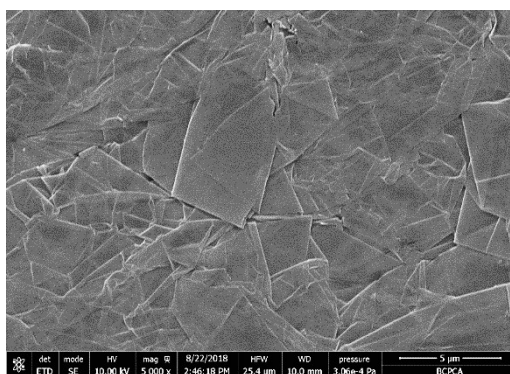


Figure S1. The SEM image of pristine graphite paper.

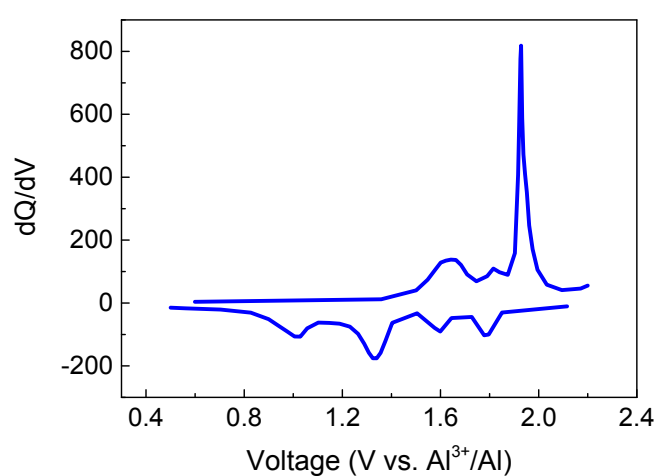


Figure S2. The dQ/dV plot of the ternary inorganic molten salt-based AIB operating at 99 °C.

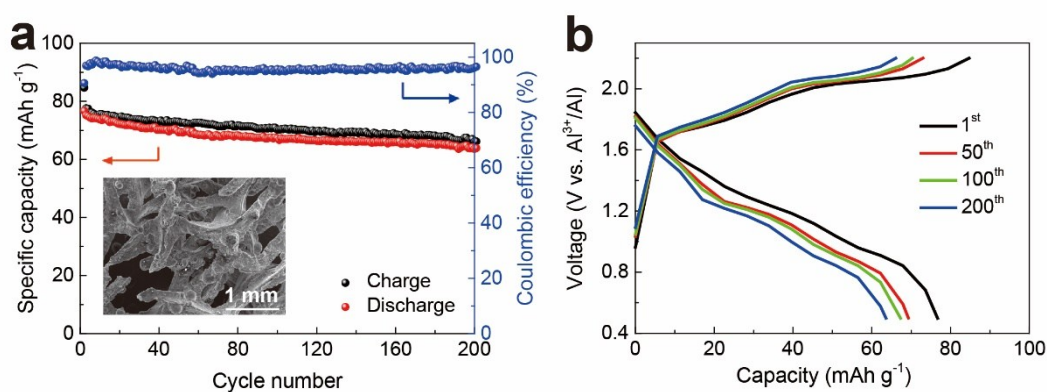


Figure S3. Electrochemical performance of the AIB using Al foam as the anode material. (a) Cycle performance of the AIB acquired at 500 mA g⁻¹ and 99 °C. The inset is an SEM image of the Al foam. (b) The corresponding charge/discharge profiles from selected cycles in (a).

A three-dimensional Al foam was used as the anode material to fabricate AIBs. Compared with Al foils, Al foam provides more pathways for electrolyte transportation, larger surface area for the interfacial electrochemical reactions, and more volume for loading other materials to prepare composites. Figure S3a displays the charge/discharge capacities of an Al foam-based AIB at 500 mA g⁻¹ for 200 cycles. A charge/discharge capacity decay from ~84.7/76.7 mAh g⁻¹ to ~66.2/63.9 mAh g⁻¹ was observed. The Columbic efficiency of the battery maintains a value of ~96 ± 1%. Figure S3b displays representative charge/discharge profiles of the battery recorded at different cycles, showing a similar feature to the Al foil-based AIBs.

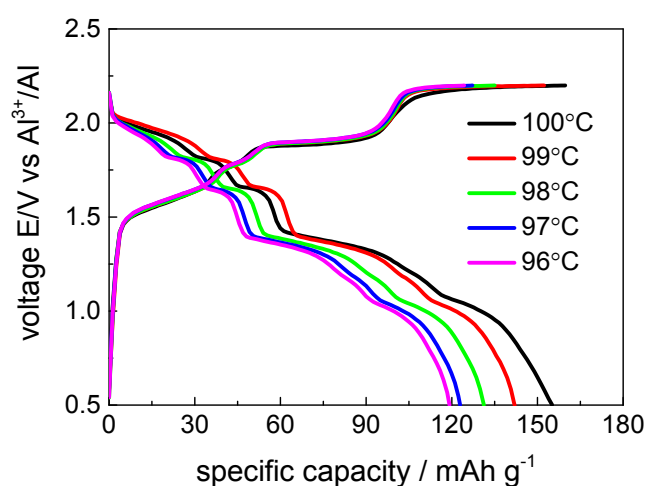


Figure S4. The typical charge/discharge profiles of AIBs at 200 mA g⁻¹ and a series of temperatures.