

Supplementary Information

A room-temperature rechargeable dual-plating lithium-aluminum battery

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Experimental Section

Materials Characterizations

X-ray diffraction (XRD) patterns were obtained by Bruker D2 Phaser X-ray diffractometer with Cu K α X-ray source. The scanning electron microscopy (SEM) images and energy dispersive spectroscopy (EDS) elemental mappings were attained with JEOL-7100F microscope.

Electrochemical Measurements

The electrochemical performance of liquid electrolyte was evaluated by assembling CR2016 coin-type cells in an argon-filled glove box, and glass fiber membrane (GF/D What-man) was used as the separator. For Li-Al battery and “cathode-free” Li-Al battery, the “H”-type electrochemical pools was employed. The lithium metal foil pressing on steel mesh was used as anode with 1 M LiPF₆ dissolved in mixture of ethylene carbonate (EC), diethyl carbonate (DEC) and dimethyl carbonate (DMC) with volumetric ratio of 1:1:1 as electrolyte in anode side. 1 M LiAlCl₄ dissolved in AlCl₃-[EMIm]Cl (1-ethyl-3-methylimidazolium chloride)

(1.3:1) was used as electrolyte in cathode side (catholyte) and $\text{Li}_{1.5}\text{Al}_{0.5}\text{Ge}_{1.5}(\text{PO}_4)_3$ ceramic was used as separator to separate the electrolytes in two sides. The $\text{Li}_{1.5}\text{Al}_{0.5}\text{Ge}_{1.5}(\text{PO}_4)_3$ ceramic separator was purchased from Hefei Kejing Materials Technology Co., Ltd., and the nominal thickness and ionic conductivity are 0.2 mm and $4.5 \times 10^{-4} \text{ S cm}^{-1}$ at room temperature, respectively. For Li-Al battery, aluminum foil was used as cathode, while aluminum foil was replaced by Mo current collector in the “cathode-free” Li-Al battery. For “cathode-free” Li-Zn battery, except for that the catholyte is changed to 1 M LiCF_3SO_3 and 0.5 M $\text{Zn}(\text{CF}_3\text{SO}_3)_2$ dissolved in acetonitrile, the other components are same with “cathode-free” Li-Al battery. Galvanostatic charge/discharge and cyclic voltammetry (CV) test was performed by using a VMP3 multichannel electrochemical workstation (Bio-Logic France).

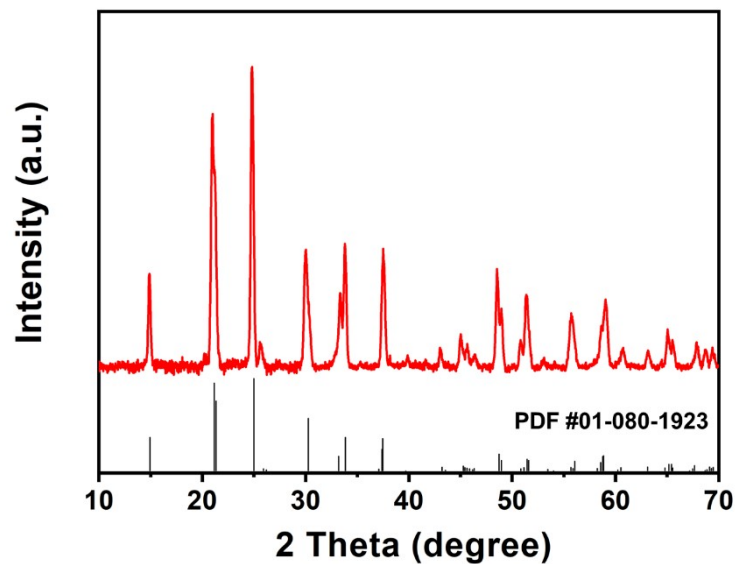


Fig. S1 XRD pattern of $\text{Li}_{1.5}\text{Al}_{0.5}\text{Ge}_{1.5}(\text{PO}_4)_3$ ceramic separator.

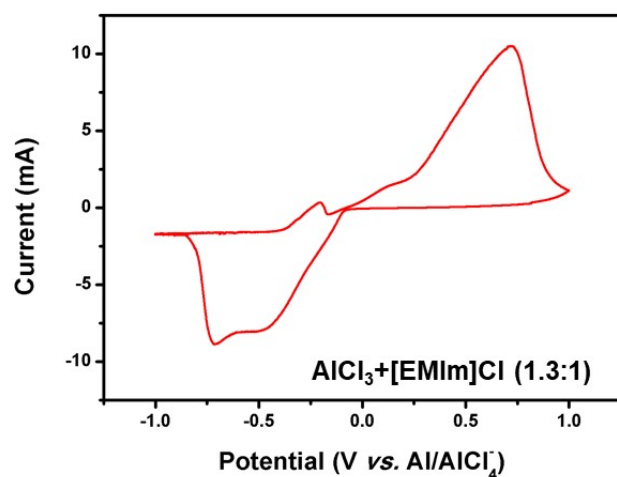


Fig. S2 CV curve of Al-Mo battery with AlCl₃-[EMIm]Cl (1.3:1) ionic liquid as electrolyte at 50 mV s⁻¹.

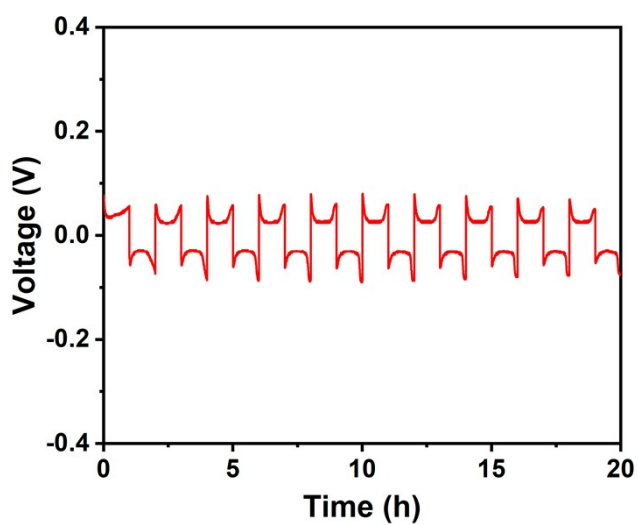


Fig. S3 The voltage-time curves of Li-Li battery with lithium organic electrolyte at 0.5 mA cm⁻² without Li_{1.5}Al_{0.5}Ge_{1.5}(PO₄)₃ ceramic separator.

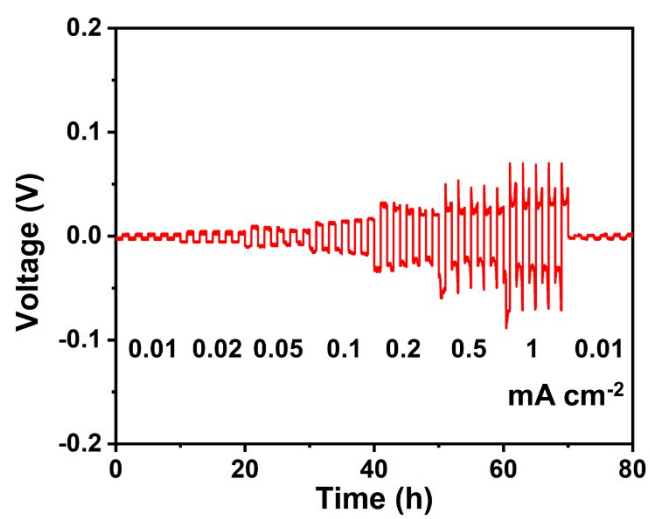


Fig. S4 The voltage-time curves of Li-Li battery with lithium organic electrolyte at different current density.

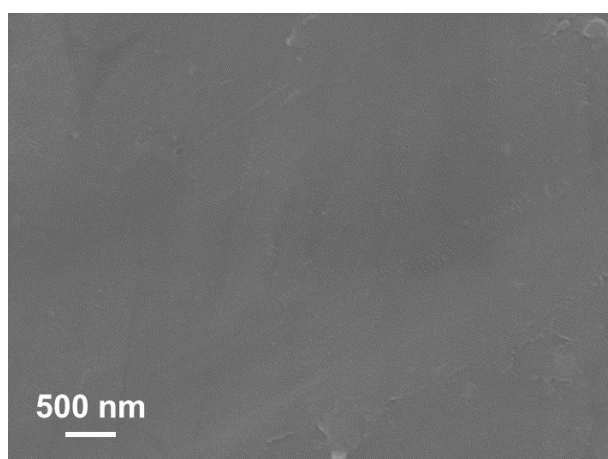


Fig. S5 SEM image of Mo current collector before plating process