

Supporting Information

A Simple Cl⁻ Free Electrolyte Based Magnesium Nitrate for Mg-sulfur Battery Applications

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Abstract The Magnesium-sulfur battery (MgS) is a promising alternative for the post-lithium battery due to its low-cost construction, eco-friendliness, high theoretical energy density, and safety. However, the lack of simple compatible electrolytes, self-discharge, polysulfide shuttle effect, and the slow conversion reaction pathway still limits its practical applications. Here, we propose a simple halogen-free electrolyte (HFE) based on Mg(NO₃)₂ dissolved in the cosolvent of acetonitrile (ACN) and tetra ethylene glycol dimethyl (G4), that applies for a Mg/S full cell. The as-prepared Mg-ion electrolyte has exhibited efficient Mg plating/stripping performance, high anodic stability (vs Mg/Mg²⁺) and the high ionic conductivity of $\sim 10^{-4}$ S cm⁻¹ at 313 K. Chronoamperometry, Scanning Electron Microscopy and Energy Dispersive Spectroscopy examinations report that the HFE supports flat, dendrite-free, and translucent Mg deposits. A polymer layer interface (PLI) based polyvinylidene fluoride (PVDF) and Mg(O₃SCF₃)₂ has been designed to isolate the surface of the Mg anode from the liquid electrolyte. The sulfur cathode with anchoring materials of silicon carbide and barium titanate-based has been designed and characterized. The Mg/S battery has constructed with an initial discharge capacity of up to 1200 mAh g⁻¹ and retained a reversible capacity at 100 mAh g⁻¹ after ten cycles. This study offers a

pivotal role in designing a promising halogen-free electrolyte candidate for a high-performance MgS battery.

Keywords: Magnesium ion battery; Halogen-free electrolyte; Sulfur; Polymer interface; Polysulfide

Table S1 Comparison between the CE values at RT and 55 °C

CVs at RT	Integrate of plating	Integrate of stripping	CE%=(Stripping/plating)*100%	CE% average
CE1	-7.78766E-5	2.62831E-5	33.75%	32.69%
CE2	-8.45777E-5	2.66583E-5	31.52%	
CE3	-8.24676E-5	2.62058E-5	31.78%	
CE4	-7.47965E-5	2.52099E-5	33.70%	
CVs at 55 °C	Integrate of plating	Integrate of stripping	CE%=(Stripping/plating)*100%	CE% average
CE1	-8.28837E-4	1.8903E-4	22.81%	23.53%
CE2	-8.13929E-4	1.88225E-4	23.13%	
CE3	-7.8328E-4	1.85912E-4	23.74%	
CE4	-7.46803E-4	1.82569E-4	24.45%	

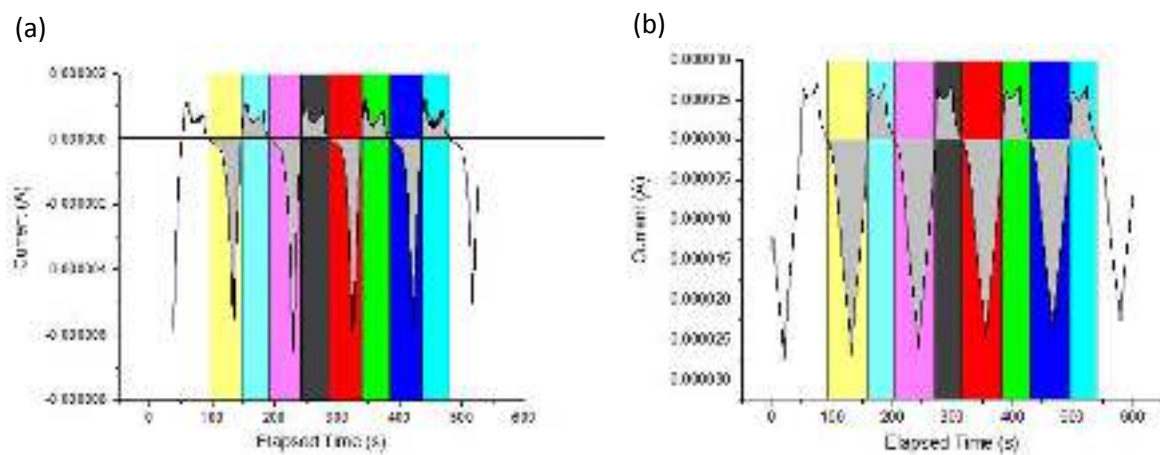


Fig. S1 CE calculation for the CVs at (a). RT (b). 55 °C

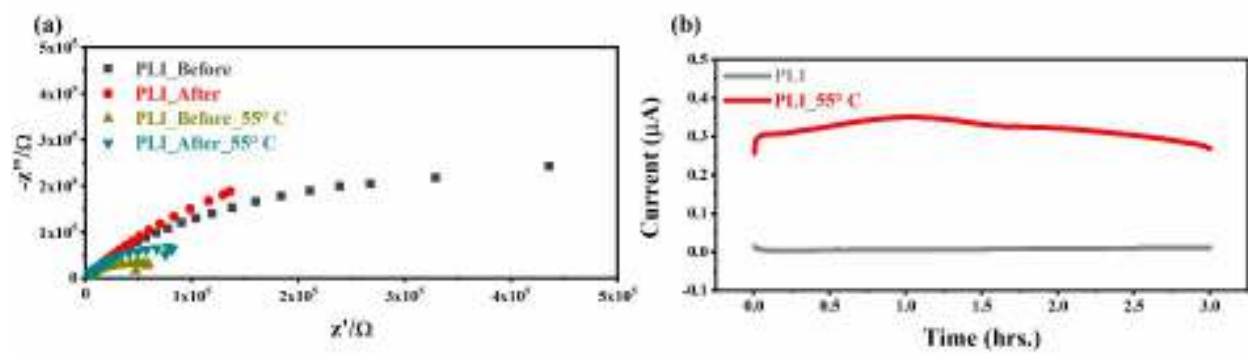


Fig. S2 (a) Nyquist plots of a Mg//PLI//Mg cell before and after DC polarization; (b) Current curve vs time at the applied voltage of 0.1 V of the symmetric Mg// PLI //Mg.

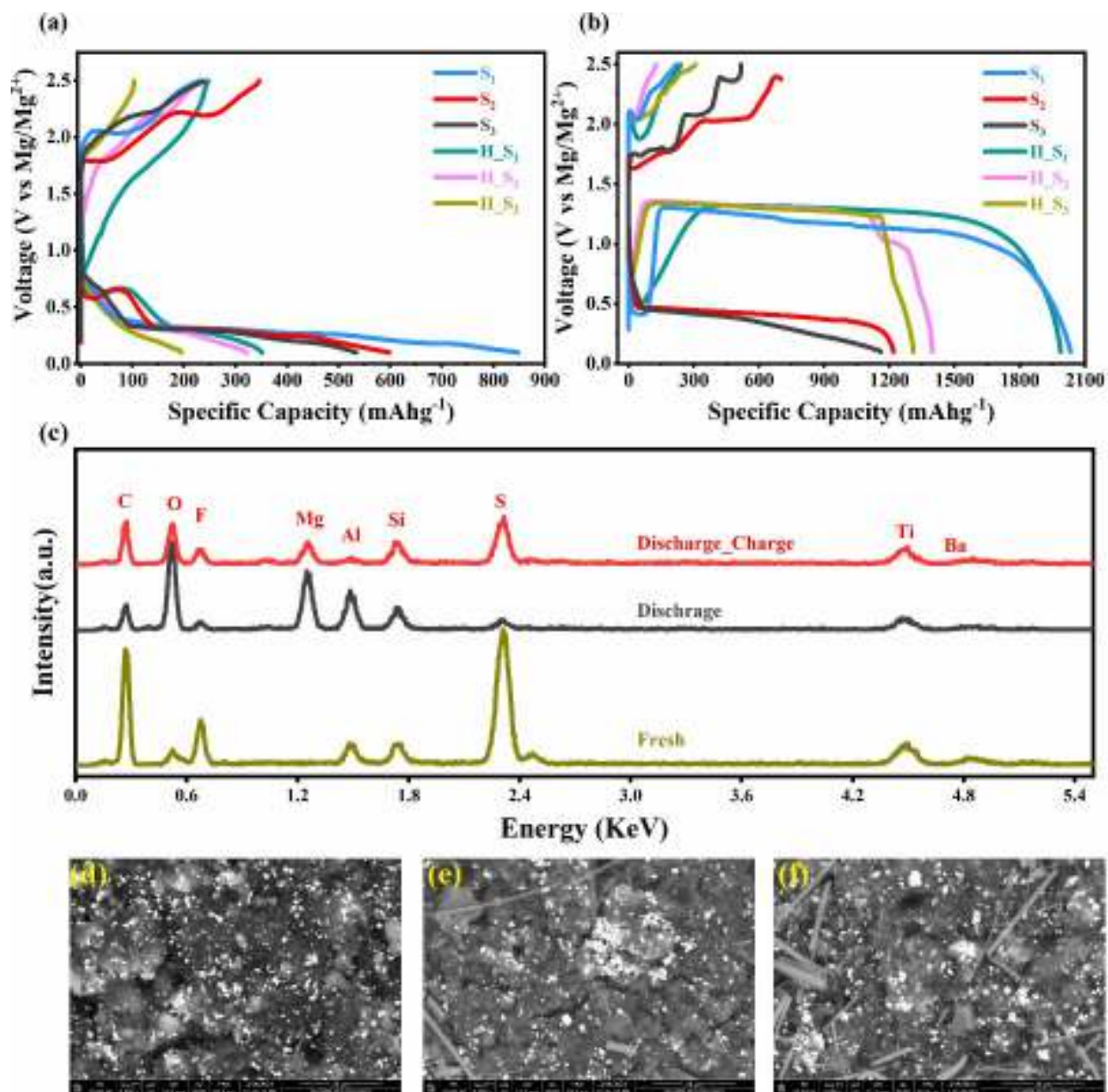


Fig. S3 Galvanostatic Discharge_Charge curves of the samples for (a) HFE at room temperature; (b) HFE at 55 °C; (c) EDS spectra of the S_3 cathode sample; SEM micrographs of the S_3 cathode sample: (d) Fresh, (e) Discharge , (f) Discharge_Charge.