4th International Symposium on
Energy Challenges & Mechanics
- working on small scales

11-13 August 2015
Aberdeen, Scotland, UK

Ether-based electrolyte for lithium metal anode in Li-Air batteries

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Accepted for publication on 28th February 2015

Lithium-air(oxygen) batteries attract attention for their higher theoretical energy densities than conventional lithium ion batteries. The battery suffers from the lithium dendrite issue that prevents the widespread utilization of lithium as an anode in practical rechargeable batteries. Poly(ethylene oxide) (PEO)-based solid electrolyte using a variety of additives have been extensively studied in our research group with the aim of prolonging the onset of lithium dendrite formation. Such lithium electrode covered with a solid electrolyte is called protected lithium electrode (PLE). To obtain high-energy density lithium-air batteries greater than 500 Wh kg^{-1}, the specific areal capacity should be more than 10 mAh cm^{-2} at the operating current density. Unfortunately, PLEs with PEO-based interlayers have fallen short of this expectation. In this study, we present lithium bis(fluorosulfonyl)imide (LiFSI) and tetraethylene glycol dimethyl ether (G4) as a novel interlayer in place of PEO_xLiTFSI to give metallic lithium anodes with high specific areal capacity of more than 10 mAh cm^{-2} in the absence of lithium dendrite formation.

A symmetrical Li/LiFSI-2G4/Li cell at 60 ºC showed no short-circuit for 40 h that corresponds to the full capacity of loaded lithium anode at 1 mA cm^{-2}. On the other hand, potential divergence was observed for the cell of Li/LiFSI-20G4/Li cell ca.12 h at 1 mA cm^{-2} and 60 ºC. Raman measurements revealed that all the solvent molecules of G4 was completely bound to lithium cations to form a solvated ionic liquid for x=1 and 2 in LiFSI-xG4. In higher x values, there are always free solvent molecules in addition to these pseudo-ionic liquids. The higher polarization in the diluted LiFSI-xG4 may be caused due to irreversible decomposition of the free G4 solvents or simply less salt concentrations. Lithium anode can deliver a high specific areal capacity of 12 mAh cm^{-2} for 40 cycles at 6 mA cm^{-2} and 60 ºC in LiFSI-2G4. We demonstrated that LiFSI-2G4 as the interlayer allows the favorable cyclability of high energy density aqueous lithium-air cell. It is believed that LiFSI-2G4 is a promising electrolyte for lithium electrode at elevated temperature.

Keywords: lithium-air battery; dendrite; tetraglyme; LiFSI