Abstract #2379, 224th ECS Meeting, © 2013 The Electrochemical Society

Self-Healing Electrostatic Shield Mechanism for Dendrite Control during Electrodeposition

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Rechargeable lithium metal batteries are considered the "holy grail" of energy storage systems. Unfortunately, uncontrollable dendritic lithium growth inherent in these batteries (upon repeated charge/discharge cycling) has prevented their practical application over the past 40 years. We show a novel mechanism which can fundamentally alter dendrite formation. At low concentrations, selected cations (such as cesium or rubidium) exhibit an effective reduction potential below the standard reduction potential of lithium ions. During lithium deposition, these additive cations form a positively-charged electrostatic shield around the initial growth tip of the protuberances without reduction and deposition of the additives. This shield forces further deposition of lithium to adjacent regions of the anode and eliminates dendrite formation in the deposited lithium film (see Fig. 1). When 1M $LiPF_6$ in PC is used as the base electrolyte, a dendrite free lithium film can be obtained by adding 0.05M $CsPF_6$ in the electrolyte. However, the Coulombic efficiency of the deposition is only ~76%. By optimizing electrolyte solvent (both solvent and salt), dendrite free lithium films have been obtained with a Coulombic efficiency of >97%. Further development of this approach will lead to long term safe operation of rechargeable Li metal batteries.

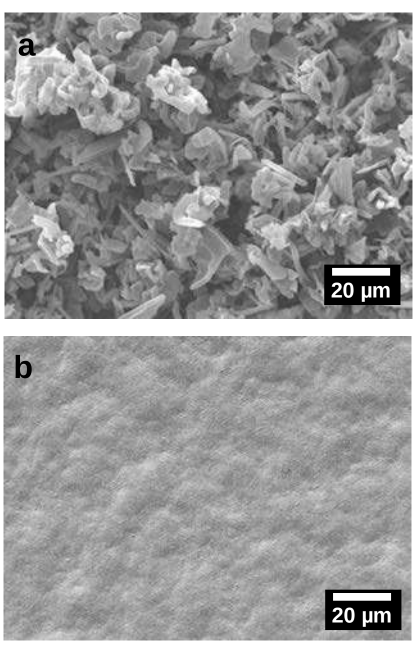

Fig. 1(a) SEM images of Li metal film grown on Cu substrates: (a) with no additive; (b) with additive.

This strategy may also prevent dendrite growth in lithium-ion batteries as well as other metal batteries and transform the surface uniformity of coatings deposited in many general electrodeposition processes. Furthermore, the fundamental scientific principles that transform a chaotic/dendritic growth pattern to an ordered/spheroidal growth pattern may also explain other naturally-occurring phenomena and provide possible guidelines to control heterogeneous nucleation and growth in other fields.