

Monitoring of Lithium Plating by Neutron Reflectometry

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Abstract

The development of high-capacity rechargeable and safe metallic lithium negative electrodes for next-generation batteries requires an in-depth understanding of reasons for nonuniform lithium plating during lithium-metal battery charge. It drives the interest for the tools enabling efficient monitoring of electrochemical interfaces where lithium electrodeposition occurs. We report on a three-electrode electrochemical cell designed to track lithium electrodeposition from aprotic electrolytes by neutron reflectometry (NR) in the specular reflectivity mode. We performed a case study of Li plating from LiClO₄ solution in propylene carbonate. The sensitivity was optimized by tuning the neutron scattering contrast for a given electrode material (Cu film) and the electrolyte, which was done employing a deuterated solvent. The analysis of the scattering length density (SLD) profiles derived from the modeling of the reflectivity data clearly demonstrated that the deposition of nm-thin Li layers above initially formed solid-electrolyte interphase (SEI) layer can be detected and their roughness, which is a characterizing parameter of electrodeposition nonuniformity, can be estimated. It makes NR a proper tool for further studies of "dendritic" lithium growth.