LIPSS for Li-less lithium metal negative electrodes of secondary aprotic batteries

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Over the past three decades, significant research and development investments from various sectors including SMEs, academia, and national laboratories have pushed the advancement of battery technology, catering to the needs of portable electronics, devices, and hybrid electric vehicles. This remarkable technological evolution hinges on lithium-ion intercalation chemistry, renowned for its remarkable versatility. However, as the lithium-ion intercalation approach is reaching its theoretical limits, there arises a pressing need for innovative methods to store more energy in batteries based on them.

In this study, we propose a novel approach employing (a) laser-induced periodic surface structure (LIPSS) patterning under ambient conditions, and (b) coating with an artificial solid-state electrolyte (aSEI) to fabricate a stainless-steel (SS) lithium-less Li metal electrode (L3ME). This electrode shows the capability to reversibly plate and strip lithium for hundreds of cycles in an aprotic galvanostatic cell. The LIPSS technique facilitates the creation of regular micrometer-long ripples on the surface of SS, with periods falling within the 150-250 nm range. Concurrently, aSEI deposition induces the formation of a smooth surface morphology through the homogeneous dispersion of a polymeric-inorganic composite film.

An array of LIPSS patterning conditions and aSEI compositions have been thoroughly analysed. Optimal L3ME materials comprise SS thin foils featuring a mesostructured surface pattern characterized by a regular distribution of ripples composed of Fe and Fe_2O_3. This structured surface pattern is hidden beneath a uniform and smooth polyethylene oxide-$LiNO_3$ composite film. Notably, L3ME electrodes exhibit superior performance in aprotic lithium cells, accommodating fully reversible metallic Li stripping and deposition with coulombic efficiencies of 100% over hundreds of cycles.

Comparative assessments against bare copper electrodes and other lithium-less substrates underscore the unique synergistic effects of LIPSS and aSEI, enhancing plating and stripping reversibility by selectively impeding electrochemical lithium dissolution.

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