Towards anodeless lithium metal negative electrodes for secondary aprotic batteries

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The transformation of the Li-ion battery value chain from linear to circular requires to address the challenge of the supply of critical raw materials (CRM) like, for example, natural graphite, the most used active materials for anodes in Li-ion batteries. In this respect the substitution of graphite anodes with anodeless substrates can simultaneously enhance circularity and improve the performance thanks to the exploitation of the lithium plating/stripping reactions using the Li^+ ions stored in the cathode. In this communication we discuss the synergistic use of metal laser patterning in air (laser induced periodic surface structures, LIPSS) and an artificial solid-state electrolyte (aSEI) to promote the electrochemical lithium deposition/dissolution on a metallic substrate. This approach allows the manufacture of an anodeless substrate constituted by a stainless-steel (SS) lithium-less Li metal electrode (L^3ME), that can reversibly deliver the plating/stripping of metallic lithium for hundreds of cycles in aprotic batteries. LIPSS manufacturing draws on the surface of SS regular micrometer-long ripples with lateral periodicity in the 150-250 nm range, whereas the aSEI deposition covers the nanostructured surface with a smooth polymeric-inorganic composite film. The optimized L^3ME electrodes show superior performance in aprotic lithium cells being able to accommodate a fully reversible metallic Li stripping/deposition with outstandingly high coulombic efficiencies approaching 100% for hundreds of cycles under a variety of galvanostatic conditions close to commercial standards (current densities in the range 0.25 -1.25 mA cm^{-2} and areal capacity limits in the range 0.5-5 mAh cm^{-2}).