



Some Directions for Performance Improvement of Li-Ion Batteries out of Usual Paths

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Recent developments at IMN will be shared on several research directions out of usual paths for performance improvement of Li-ion batteries. We will focus on innovative surface modifications of electrode components, new electrode compositions and architectures, and failure mechanism upon cycling by in-depth characterization through coupled advanced spectroscopic techniques. A molecular grafting approach has been proposed as a way to modify the interfacial chemical reactivity of oxide materials, which is detrimental to their long-term energy storage properties as electrodes of Li-ion batteries. Surface derivatization of powder oxide materials such as $\text{Li}_{1.2}\text{V}_3\text{O}_8$ and $\text{Li}(\text{Mn},\text{Ni})_2\text{O}_4$ was accomplished by in situ electrografting of a diazonium salt during Li-ion intercalation, leading to a covalently bonded organic multilayer. Charge transfer is not impeded, while electrolyte decomposition is inhibited thus increasing the cycle life and decreasing the self-discharge. Carbon additives of classical porous electrodes occupy a large volume fraction which is lost for charge storage. Redox functionalization of the surface of some carbon additives has been successfully achieved through non-covalent grafting chemistry using multi-redox pyrene molecules synthesized on purpose. Such functionalized carbon additives have been used to increase the stored energy and power of C-coated LFP porous electrodes. Thicker electrodes are needed for higher energy density Li-ion batteries. We evaluate different directions in order to design new innovative electrode architectures for such a purpose. Our grafting chemistry has been further developed to achieve molecular junctions between non-carbon-coated LFP and multiwall carbon nanotubes (MWCNT) using a designed thiophene-based conjugated molecule. The strategy enables original architecturing of the cathode of Li-ion batteries, with the individual MWCNT being electronically nanocontacted at the surface of LFP grains. This advancement leads to much higher specific capacity and better capacity retention for non calendared thick electrodes, for which the electronic wiring of the electroactive material grains is a critical issue. Another direction followed is the use of conducting polymer additives in porous electrodes, which are able to act as both conducting fillers and mechanical reinforcement materials. We have synthesized a new form of lithium doped PANI, the excellent properties of which in terms of specific capacity, stability on cycling and rate capability will be presented. The coating of bare LFP particles with thin layers of this new Li-doped PANI allows surpassing the performance of commercial carbon coated LFP thick electrodes. The role of this PANI additive into millimetric thick electrodes of NMC material will also be discussed. Future developments of higher energy density Si-based Li-ion batteries depend on the mastering of side reactions at the Si anode. We will compare the SEI composition and morphology at the Si surface upon cycling in half cell and full Li-ion cell configurations using a combination of ^7Li , ^{19}F MAS NMR, XPS, TOF-SIMS and STEM-EELS. The origin of the much faster aging of Si-based full cells versus half cells and future directions for improvement will be discussed

Résumé en anglais

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