**Optimizing the water-soluble binder with various additives for Li4Ti5O12 lithium-ion anodes**

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Environmentally friendly Li4Ti5O12 (LTO) is considered the anode material of choice for low-cost, high-power lithium-ion batteries. Aqueous processing of LTO electrodes is in the need of cost saving and preservation of the environment, we developed an optimized combination of processing additives with sodium carboxymethyl cellulose (CMC) binder to ensure high-performance lithium storage. Interestingly, certain compositions reveal the growth of new particles upon electrode processing, for which the electron transport appears to play a decisive role. Accordingly, the structure evolution of LTO particles in aqueous processing, such as lattice parameter, unit cell volume, μstrain and oxygen position, was first time to be investigated through analysis of *in-situ* XRD patterns. Besides, the surface morphology properties must be controlled due to influence on the electrochemical performance of the materials. Indeed, the ball milling speed and CMC percentage on the electrode formulation have been proved to be the crucial factors that influence the surface morphology of LTO electrodes during aqueous-based electrode preparation. In fact, less amount of CMC binder in the final electrode formulation decreases the viscosity of the slurry and the tunnels resistances related to the gap between the binders and the particle clusters. Therefore, controlled surface morphology, as well as, electrode formulation, is beneficial for enhancing the cycling capability of LTO anode material for Lithium ion batteries.

Li3PO4 could be detected in theXRD pattern upon the addition of additive during aqueous preparation of LTO electrodes, however, this additive had detrimental impact on the electrode adhesion. Therefore, we had directly tried Li3PO4 as additive to prepare electrodes, luckily, high mass loading electrodes were available in the favor of carbon-coated aluminum foil and modified solvent. We also combined two additives together to reduce the cell polarization and improve the lithium-ion diffusion and transfer kinetics in LTO electrodes, and the interaction of two additives, the impact of additives on CMC binder and LTO was further investigated by FTIR, EIS, CV and GITT. It is worthwhile to mention that ball milling speed also has an effect on the evolution of the slurry pH value, lattice distance and Li+ location variation in LTO structure. Finally, the electrolyte formulation was also modified by adding FEC as additive, which leads to the formation of a thicker SEI layer on the surface of LTO electrodes and the enhanced cycling stability in both half- and full-cell at 2C rate.