Analytical multi-probe study of the SEI on silicon based electrode in full cell configuration

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Over the past years, lithium-ion battery became one of the most promising energy storage technology addressing a large range of applications, from hybrid electric vehicles, electric vehicles to Plug-in Hybrid Electric Vehicles, which are essential to reduce the fossil oil dependency. In order to meet requirements of these automotive applications, it is necessary to find both higher capacity and higher capacity retention electrode materials for Li-ion batteries. Providing new materials for the negative electrode of Li-ion cells has therefore been the subject of many investigations for more than a decade. Among the candidates for negative electrode, silicon appears as an attractive alternative to graphite due to its natural abundance, high specific gravimetric capacity (3579 mAh.g^{-1} vs. 372mAh.g^{-1} for graphite) and a large volumetric capacity (2081 mAh.cm^{-3} vs. 779 mAh.cm^{-3}). Silicon based electrodes however suffer from poor cyclability due to the large volumetric expansion of Si particles upon cycling as well as an unstable solid electrolyte interphase (SEI) caused by a continuous liquid electrolyte degradation at the surface of the Si phase leading to the increase of the amount of electrolyte-degradation products. Although the interface between Si particles and the electrolyte is playing a major role in the electrochemical performance, it has rarely been characterized in depth and the failure mechanism of silicon-based electrodes has been studied only in a half-cell configuration. In the present work, a combination of analytical techniques, ^{7}Li, ^{19}F MAS NMR, XPS, TOF-SIMS and STEM-EELS, provides an in-depth characterization of the SEI forming on the surface of silicon particles as well as its evolution upon cycling in a full Li-ion cell configuration with LiNi$_{1/3}$Mn$_{1/3}$Co$_{1/3}$O$_2$ as the positive electrode.