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The influence of counter electrode on the capacity fading in LiNi$_{0.33}$Mn$_{0.33}$Co$_{0.33}$O$_2$-based Li-ion battery cells

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In commercial Li-ion cells, LiNi$_{0.33}$Mn$_{0.33}$Co$_{0.33}$O$_2$ (NMC) is a common positive electrode material rendering higher energy density than materials such as LiMn$_2$O$_4$ (LMO) and LiFePO$_4$ (LFP) and being cheaper than LiCoO$_2$ (LCO). In this study, NMC-based cells with different common negative electrode materials are compared in order to evaluate how the capacity fading changes. The negative electrode materials used are lithium foil, graphite and Li$_4$Ti$_5$O$_{12}$ (LTO). Lithium foil is known for its high energy density, but has safety problems preventing it from being used in most commercial applications. Graphite is the most common material used in Li-ion batteries as negative electrode. It has a decent capacity compared to many other electrode materials and a low operating potential, which is beneficial for energy density. However, the low operating potential of graphite and lithium foil cause reduction of the electrolyte. To avoid this problem LTO can be used, operating at a potential at which the electrolyte is stable. The high operating potential decreases the energy density in LTO-based cells, but it has a high rate capability making it a good solution in applications where high power is required.

It is observed that a rapid capacity fade takes place in cells where lithium foil is used, mainly attributed to the increase in over-potential as the cycling time increases, see Fig. 1. The increased over-potential is either caused due to a thicker surface layer on the lithium foil or the NMC electrode, as the bulk material remains stable according to x-ray absorption near edge structure (XANES) spectra. The thickness of the surface layer on the NMC electrode increases during cycling, supporting that electrolyte degradation products is the cause for the increased over-potential. The surface layer also to a large extent consists of organic compounds, as compared to when LTO or graphite is used. For cells containing graphite and LTO electrodes the capacity fading is slow. Also the over-potential in these cells remain at a stable low value throughout the cycling, indicating more stable electrode surface layers.

![Figure 1. Potential profiles of NMC electrodes cycled versus different negative electrodes. a) NMC vs. Li-metal. b) NMC vs. LTO. c) NMC vs. graphite.](image-url)