# Mitigating the Impedance Growth in SiG//NMC811 Lithium-Ion Batteries by Prelithiation of the Silicon-Graphite Anode

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Lithium-ion batteries, consisting of a silicon-graphite (SiG) anode and a $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$ (NMC811) cathode offer the potential of high cell-level energy densities >300 Wh $kg^{-1}$ [1]. However, the realization of these theoretical values continues to pose a significant challenge. The two major obstacles include (i) the ongoing consumption of active lithium caused by side reactions at the silicon-electrolyte interface [2], and (ii) the limitation of the end-of-charge potential of the NMC811 cathode due to the release of lattice oxygen and subsequent capacity fading [3].

In the present work, we investigate the impact of the upper cell cutoff voltage on the cycling stability and the lithium inventory of SiG//NMC811 full-cells at 45°C. Hence, we first benchmark the NMC811 cathodes (areal loading of ~2.0 mAh $cm^{-2}$ at 4.1 V vs. $Li/Li^+$) in half-cells against lithium metal anodes, by applying upper cutoff voltages between 4.0 and 4.6 V vs. $Li/Li^+$. Further, we investigate SiG//NMC811 full-cells, utilizing a capacitively oversized SiG anode (~7.0 mAh $cm^{-2}$), in terms of their cycling stability and electrode polarization at different C-rates between 0.1 and 1.0 $h^{-1}$. Using a gold-wire micro-reference electrode we monitor the potentials and the impedance growth of the individual electrodes as a function of the cycle number and the upper cell cutoff voltage, respectively. Finally, we complement the impedance measurements by an *operando* gas evolution analysis of the NMC811 cathode using on-line electrochemical mass spectrometry (OEMS) [3].

Our results indicate that the relatively high lithiation potential of the capacitively oversized SiG anode causes a gradual upward shift of the end-of-charge potential of the NMC811 cathode, which leads to the release of NMC811 lattice oxygen and a concomitant impedance growth at the positive electrode, even at comparatively low cell cutoff voltages. To cope with this challenge, we demonstrate that *ex-situ* prelithiation of SiG anodes represents an effective strategy to reduce the average anode potential and thus mitigate an upward shift of the end-of-charge potential of the NMC811 cathode, while additionally providing a sufficiently large lithium inventory to allow a reversible capacity of >180 mAh $g^{-1}_{NMC}$ after 250 cycles at 45°C. We conclude our analysis with a critical review on the effective energy densities that can be realized with the SiG//NMC811 cell chemistry and compare it to the practical energy density of state-of-the-art graphite//NMC622 lithium-ion batteries.

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**References:**

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