

# **The Spontaneous Intercalation of Lithium Metal into Graphite Electrode – A mechanistic study.**

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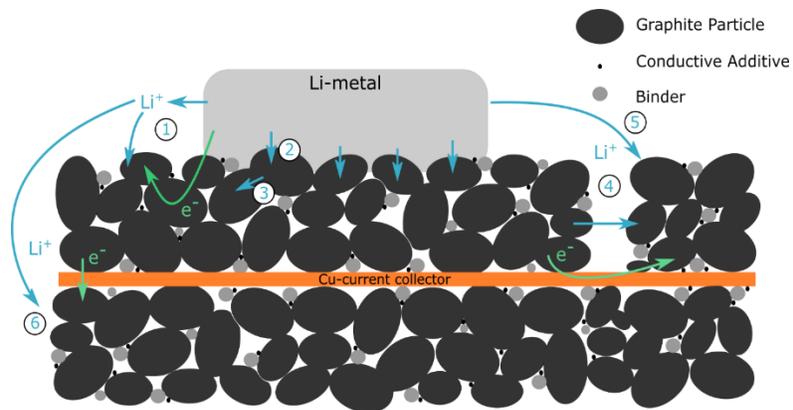
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Lithium deposition on negative graphite electrodes is one of the main aging mechanisms of Lithium-ion cells [1]. When Lithium is deposited, it spontaneously intercalates into graphite, changing the local charge distribution of the electrode [2,3,4]. Inhomogeneous charge distribution leads to an accelerated aging of the Lithium-ion batteries [2]. However, there is a lack of knowledge on the involved mechanisms of Lithium re-intercalation, although they are of high relevance.

In order to gain a better understanding of the spontaneous re-intercalation, re-distribution and relaxation processes of Lithium within porous graphite electrodes, we conducted model experiments with Lithium metal pressed on graphite surfaces. The mixed open circuit potential (OCP) of the graphite/Lithium electrode was measured in pouch half-cells during the re-intercalation. The charge distribution within the graphite electrode was investigated with Raman spectroscopy, glow-discharge optical-emission-spectroscopy (GD-OES), and optical microscopy (OM). Additionally, simulations using the pouch half-cell set-up were conducted to gain time-dependent information on charge distribution in both electrode and electrolyte. The electrolyte was systematically varied in these experiments, which revealed new insights on the pathways of Lithium-ions (see Figure). For example, a solid-state reaction between Lithium metal and graphite is possible even without electrolyte, however, the reaction using ionic pathways is much faster if both electrolyte and conductive salt are present. The presence of a counter electrode can also influence the reaction pathways. Conclusions are drawn on post-mortem methodology, inhomogeneity of aging mechanisms, and pre-lithiation of graphite electrodes.



References:

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- [2] Forouzan et al. *Journal of The Electrochemical Society* 165 (2018) A2127.
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- [4] Lüders et al., *Journal of Power Sources* 342 (2017) 17.