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# Charge Relaxation within Silicon/Graphite Anodes – A Multi-Method Study

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As silicon/graphite (SiG) composites are more commonly used as the anode active material in commercial Li-ion batteries,1 the importance of investigations of the (de-)lithiation behavior of the blended anodes grows. In this study, the charge redistribution between graphite and silicon was investigated in graphite-NMC 622 and SiG (~20 wt.-% Si) –NMC 622 bilayer pouch cells using in situ and operando X-ray diffraction (XRD). In addition, ex situ and in situ optical microscopy (IOM), as well as 3D microstructural resolved simulations employing digital twins of the cells, were carried out. Different SOC values (0%, 25%, 50%, 75%, and 100%) and two different C-rates (0.1C and 0.5C) were compared in cells during operation and in the relaxed state. Insights into the relaxation process at 75% SOC were gained by tracking of the charge redistribution in IOM cells. Ex situ optical microscopy measurements reinforced the findings of the IOM measurements. Both XRD and optical microscopy showed the disappearance of Li in the graphite component of the SiG anode during the relaxation period (≥24h) at SOC ≤75%, indicating a redistribution of Li from graphite into Si in the anode. The simulations allowed a detailed analysis of the Li concentration in both active material components during charge and relaxation, verifying the observations from the XRD and microscopy experiments. The gained insights can support a better understanding of aging of blended SiG anodes during operation.

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