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Charging and Aging Mechanisms of Si/C Composite Anodes –An Operando Neutron Scattering Study

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Interesting candidates to fulfill the requirement of higher specific energy in Li-ion batteries are Si/C composite anodes. We investigated the charging mechanisms in commercial 18650 cells (anode: graphite + 3% Si, cathode: NCA)¹ via operando neutron diffraction with the STRESS-SPEC instrument at the neutron source FRM II in Garching.

Although the amorphous Si compound did not allow following the Si alloying directly, it can indirectly be observed by the relaxation of the LiC₆ and LiC₁₂ reflexes, similar like for Li deposition.² Charging at 0.1C, 0.5C, and 0.75C showed different mechanisms. Interestingly, we find a transfer of Li from graphite into the Si compound which was, to best of our knowledge, not reported before. The measurements are complemented with simultaneous electrochemical measurements and Post-Mortem analysis (SEM and GD-OES depth profiling¹) after cell disassembly.

The unique capabilities of the used methods, especially operando neutron diffraction at FRM II give very interesting novel insights into battery materials, which will allow improvement of later battery generations.

References:

1. K. Richter et al., J. Electrochem. Soc. 165 (2018) A3602.
2. V. Zinth et al., J. Power Sources 271 (2014) 152.

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This work is based upon experiments performed at the STRESS-SPEC instrument operated by FRM II at the Heinz Maier-Leibnitz Zentrum (MLZ), Garching, Germany.

Primary authors: Dr WALDMANN, Thomas (ZSW –Zentrum für Sonnenenergie- und Wasserstoff-Forschung Baden-Württemberg); Mr RICHTER, Karsten (ZSW –Zentrum für Sonnenenergie- und Wasserstoff-Forschung Baden-Württemberg)

Co-authors: Dr PAUL, Neelima (Technical University of Munich, Heinz Maier-Leibnitz Zentrum (MLZ)); Mr NICOLA, Jobst (ZSW –Zentrum für Sonnenenergie- und Wasserstoff-Forschung Baden-Württemberg); Dr GEORGE SCURTU, Rares (ZSW –Zentrum für Sonnenenergie- und Wasserstoff-Forschung Baden-Württemberg); Dr HOFMANN, Michael; Dr GILLES, Ralph; Dr WOHLFAHRT-MEHRENS, Margret (ZSW –Zentrum für Sonnenenergie- und Wasserstoff-Forschung Baden-Württemberg)

Presenter: Dr WALDMANN, Thomas (ZSW –Zentrum für Sonnenenergie- und Wasserstoff-Forschung Baden-Württemberg)

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