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Low-temperature performance of Li-ion batteries probed by high-resolution neutron diffraction and electrochemistry

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Due to high power/energy density, good cycle life and excellent storage characteristics lithium ion batteries are considered as the major energy storage technology in the field of portable electronics and electric vehicles. Despite the overall success of Li-ion energy storage technology a number of research targets toward even higher energy/power density, lighter weight, safer and lower cost rechargeable batteries has been defined. Besides these factors, having undoubtedly high scientific and technological relevance, the stability of the Li-ion cells in a broad range of environmental conditions has to be addressed, where operating temperature is the most important criteria. According to the recommended range of Li-ion battery operation (typically -20-60℃ for discharge and 0-60℃ for charge) their use in countries with moderate and frigid climate in winter time becomes quite bounded. This becomes even more critical in aerospace applications, where stable energy storage and conversion is demanded over a long period of time under even more challenging environmental conditions. Therefore broadening the temperature range where lithium ion battery technology shows stable performance and characteristics is an emergent research task. In this context the lower temperature boundary of operation has obviously higher priority compared to the high temperature one.

The poor performance of Li-ion cells is usually ascribed to effects like low electrolyte conductivity, slow kinetics of charge transfer, increase of the solid-electrolyte interphase resistance or slowing down of lithium diffusion through it, lithium plating or to a combination of these different factors. Active research devoted to boost electrolyte performance further below the freezing point and towards higher conductivity. In the current contribution a combined in situ neutron powder diffraction and electrochemical study on Li-ion cells of 18650-type, in a temperature range -43-37℃ is reported. The main focus is put on the role of the graphite anode (as a most likely source of failure) with respect to the low temperature performance of the cell. Instead of a quasi-continuous behaviour observed at ambient temperatures, a thermodynamic anomaly occurs at the graphite anode upon a discharge at low temperature, which is primarily reflected in the character of the LiC12–to–graphite phase transformation and the unusual temperature dependence of LiC6. Freezing of the liquid electrolyte at temperatures below -23℃ in a long range ordered structure was experimentally determined with no hints for lithium plating observed [1]. Obtained anomalous behavior is associated to a thermodynamical instability of lithiated graphite phases at temperatures below -23℃, which influences the performance of Li-ion batteries at low temperatures.

[1] A. Senyshyn, M.J. Mühlbauer, O. Dolotko, H. Ehrenberg, Low-temperature performance of Li-ion batteries: The behavior of lithiated graphite, J. Power Sources 282 (2015) 235-240

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