Insights into the Degradation Mechanism of Lithium Ion Batteries by in-operando NMR Spectroscopy

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Abstract: Lithium ion batteries (LIBs) play more and more important roles in energy storage systems for a variety of applications such as mobile devices, portable tools, electric vehicles and large-scale electric power storage.[1] Elucidating the mechanisms and kinetics of electrochemical reactions during cycling, especially understanding the degradation mechanisms of LIBs is crucial for a systematic improvement of performance, lifetime and safety of LIBs. [2]

Here, we combine in-operando solid state nuclear magnetic resonance (ssNMR) with diffusion NMR to characterize the reaction and degradation mechanisms of LIBs. First, an in-operando NMR probehead and a cylindrical battery, which are suitable to for long term battery operation over several hundred cycles, were designed and prepared.[3] Secondly, different factors, such as temperature, current rate, lithium dendrite growth, electrolyte decomposition and solid-electrolyte-interphase (SEI) layer formation, that influence the electrochemical performance of $LiCoO_2$/graphite batteries were investigated. The experimental results showed that: (i) a concentration gradient of lithium ions in the electrolyte forms and the diffusivity of lithium ions in the electrolyte will decrease when the battery voltage is above 3.5 V; (ii) at low temperatures, for example at 2° C, the capacity retention decays very fast during cycling and lithium dendrite formation occurs within 100 cycles, while the decompose reaction of electrolyte are less than that at room temperature or at elevated temperature; (iii) fast charging causes capacity loss and accelerates the process of lithium dendrite growth; (iv) lithium dendrite formation, rather than electrolyte decomposition or SEI layer effects, is the main reason for the capacity degradation of a $LiCoO_2$/graphite battery. In summary, in-operando NMR spectroscopy is demonstrated to reveal the degradation mechanism of LIBs during long time cycling.

References:

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