Predicting and Extending the Lifetime of Li-ion Cells

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The use of electrolyte has been shown to have dramatic benefits to the performance and lifetime of lithium-ion cells [1-3]. However it is becoming increasingly important to understand if an additive is beneficial in a relatively short experiment as the lifetime of cells continues to increases. Figure 1 shows two types of failure mechanisms of Li[NiMnCo]O_2/graphite cells containing different electrolyte additives.

Figure 1a shows cells have been studied through the use of High Precision Coulometry [3-6] showing that short term measurements can reliably predict those cells which will show less capacity loss during long term cycling. However, with these types of cells it becomes clear which cells are better after ~50 cycles while cycling at C/10. Figure 1b shows cells that show "roll-over" type failure where almost no capacity is lost until the cells show rapid capacity fade. For these types of cells measuring the capacity loss is a hopeless method in attempting to differentiate cells without cycling all cells until failure.

Cells that were known to show this type of "roll-over" failure were manufactured with different electrolyte additives and studied with cycling on the High Precision Charger [7], storage experiments on automatic high precision storage/cycling equipment [8] and impedance spectroscopy before cycling cells until failure. Figure 2 shows that the cycles until failure (defined by capacity reaching 1.6Ah in nominally 2Ah 18650 sized cells) can be well predicted using a combination of high precision coulometry measurements and measurements of the charge transfer impedance.

This is the first time that any type of predictive model has been able to anticipate the failure of cells that show this time of "roll-over" behavior. The measurements of coulombic efficiency and charge transfer impedance were conducted over roughly 3 weeks while the cycling took months to complete. Therefore this is an excellent and reliable technique for screening additives and evaluating cell performance in a short time. This talk will discuss the experiment in more detail and the proposed model for failure and show the benefit that additives can provide to these cells.

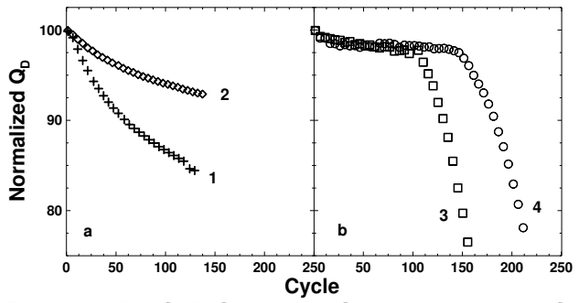

Figure 1. Panel a) shows typical capacity versus cycle number data for cells that show gradual fade over time. Panel b) shows capacity versus cycle number data for cells that show almost no capacity loss followed by "roll-over" type failure.

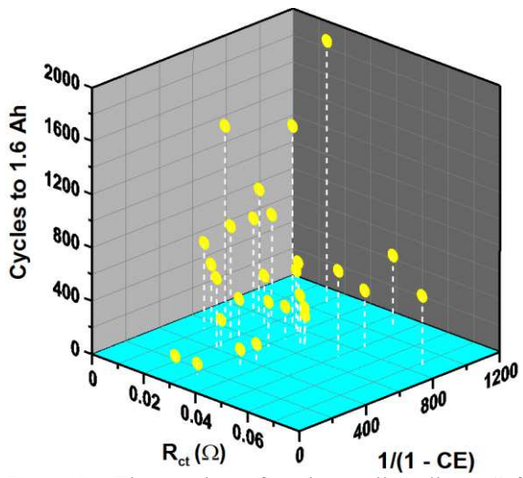

Figure 2. The number of cycles until "roll-over" failure of cells containing different additive combinations as a function of charge transfer resistance and coulombic efficiency (1/(1-CE)) showing the predictive power of short term measurements for long term performance.

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