

“Degradation of Li-ion Batteries: Experiments and Mathematical Modeling”

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Abstract

Li-ion batteries (LiBs) are the foremost candidates for powering electric/hybrid vehicles (EV/HEVs). For a LiB to be suitable for electric transportation, proper performance in terms of power and energy is required over long periods (i.e., 10+ and 15+ years for EV and HEV, respectively). In practice, unwanted aging phenomena lead to a decrease of the battery performance over time, which practically limits its lifetime. Loss of electrode active material and cyclable lithium along with the increase in cell resistance are among the most common outcomes of degradation phenomena encountered in LiBs. Although the evaluation of the battery performance under conditions typical of the end-application is crucial, an EV/HEV battery experiences a wide range of operating conditions, and as a consequence the experimental study of the battery aging in all of these conditions is impractical. In practice, one is left with trying to predict the battery life from aging studies of limited duration and based on a limited set of operating conditions.

In this presentation, a methodology for life prediction based on a physics-based model is presented and applied to commercial 2.3 Ah graphite/LiFePO₄ (LFP) batteries. First, typical aging experiments are described, along with intermediate nonintrusive checkups and postmortem analyses. The methodology does not require a specific design of experiments, and therefore experiments consist of any type of cycling and storage condition, where parameters such as current density (cycling), state of charge (storage), and temperature vary from one experiment to the other.

The aging data collected at the different checkups are then analyzed using a mathematical model that does not contain aging phenomena. This analysis allows for identifying and quantifying the different aging sources in the cell at all experimental conditions and all times.

With the information gathered from this analysis, along with the input from postmortem analyses, it is possible to include aging phenomena in the initial model and make it predictive. In our case study, aging phenomena include side reactions that lead to the growth of a passive film at the solid/liquid interface of both electrodes as well as active-material loss at the negative electrode. Ideally, a physical description of each aging phenomenon allows deriving meaningful governing equations. However, empirical correlations can be implemented into the model whenever a physical description is neither possible nor available. Examples of the life-prediction capability of the aging model are provided. The advantage of this methodology is that no extrapolation in time is needed.

In the last part of the talk, we comment on how this class of models can be further developed by resorting to “model” experiments. In such experiments, the design is simplified and operating conditions are well-controlled in order to focus on a single source of aging. The aim is to unravel poorly understood aging mechanisms and derive a meaningful set of governing equations that can be implemented in the aging model. The contamination of the negative electrode by transition metal dissolving from the positive electrode is taken as an example.

Biography

Charles Delacourt received his M. Sc. degree in analytical chemistry and electrochemistry from University Pierre & Marie Curie (Paris VI, France) in 2002, and completed his PhD in materials chemistry at Laboratoire de Réactivité et de Chimie des Solides (Université de Picardie Jules Verne, Amiens) in 2005. After a two-year postdoc within Prof. Newman group at Lawrence Berkeley National Lab and University of California, Berkeley, he has been acting as a CNRS researcher at Laboratoire de Réactivité et Chimie des Solides since fall 2007. His current research interest is the development of physics-based mathematical models for studying lithium-ion batteries, with a focus on degradation phenomena and battery life prediction. In 2012, he returned to Lawrence Berkeley Lab for a sabbatical in the group of Dr. Venkat Srinivasan, the head of the US DOE BATT program. Charles Delacourt is the author (or coauthor) of 37 peer-reviewed journal papers (among which 2 in Nature Materials), 2 patents, and is the recipient of the 2005 Research Student Award of the Battery Division of the Electrochemical Society, the 2007 Umicore Scientific Award, the 2009 Oronzio and Niccolò De Nora Foundation Prize of ISE on Applied Electrochemistry, and the 2011 Carl Wagner Medal of Excellence in Electrochemical Engineering of the European Federation of Chemical Engineering.