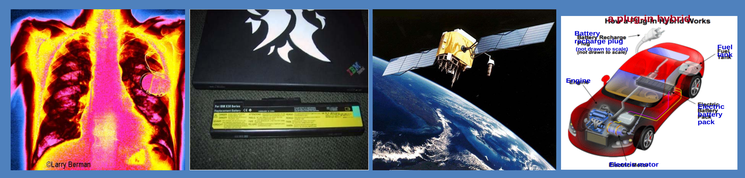
Identification of Dominant Mechanisms for Capacity Fade of Lithium-Ion Batteries

Nancy A. Burns*, Ruthvik Basavaraj**, Venkatasailanathan Ramadesigan***, Folarin Latinwo**, Ravi N. Methekar***, Richard D. Braatz**, and Venkat Subramanian***

*Department of Chemical Engineering, Tennessee Technological University, Cookeville, TN
** Department of Chemical and Biomolecular Engineering, University of Illinois at Urbana-Champaign, Urbana, IL
*** Energy, Environmental and Chemical Engineering, Washington University, St. Louis, MO

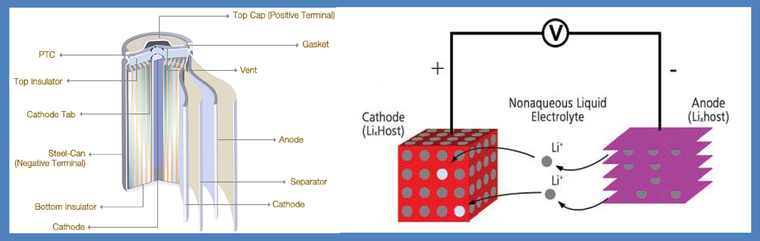
Motivation

Electrochemical power sources are expected to play a vital role in future applications of automobiles, power storage, military, mobile applications, and space. Lithium-ion chemistry has been identified as the preferred candidate for high-power/high-energy secondary batteries and commercial batteries of up to 75 Ah have been manufactured. Applications for batteries range from implantable cardiovascular defibrillators (ICDs) operating at 10 µA current to hybrid vehicles requiring pulses of 100 A.



Introduction

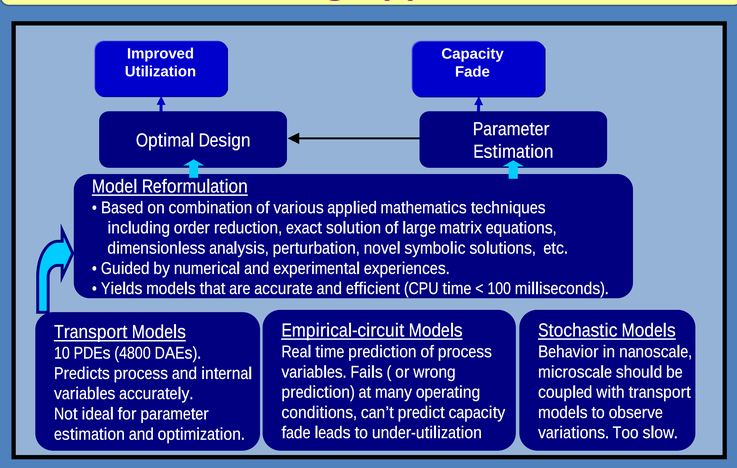
Lithium-ion battery, chemistry and reactions



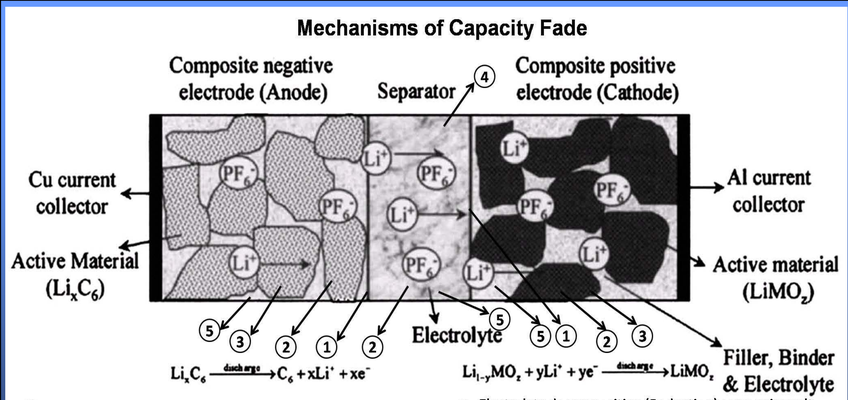
$$Li_xCoO_2 \rightleftharpoons Li_{1-x}CoO_2 + xLi^+ + xe^-$$

$$xLi^+ + xe^- + 6C \rightleftharpoons Li_xC_6$$

Modeling Approaches



Capacity Fade Mechanisms



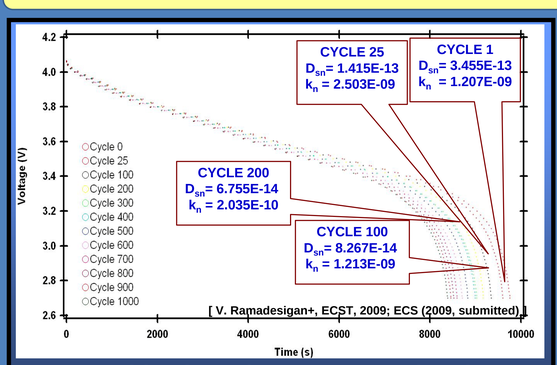
Parameter Estimation - Techniques

- **Gauss Newton Method**
  - Based on Jacobian calculation
  - Iterative procedure
  - Good initial guess is needed
- **Bayesian Estimation**
  - No need for Jacobian
  - Uncertainty analysis – Markov Chain Monte Carlo method, polynomial chaos expansions

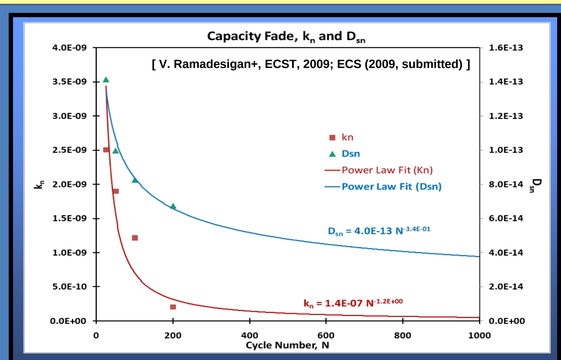
Mathematical Modeling of Capacity Fade

- Side Reactions: Represented by additional kinetic expressions, SEI layer growth: Rate of film resistance increase
- Tracking parameters with cycles (Discrete Empirical Approach)
  - Track the variations in kinetic and transport parameters with cycle number

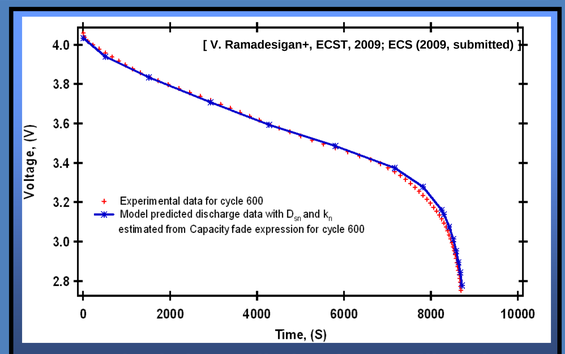
Nonlinear Estimation Results (Jacobian based)



Jacobian based parameter estimation

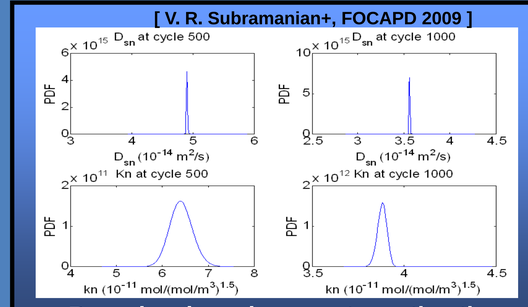


Capacity fade analysis for Quallion cells

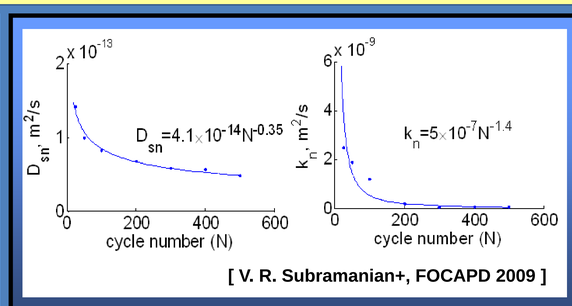


Capacity fade prediction for Quallion data

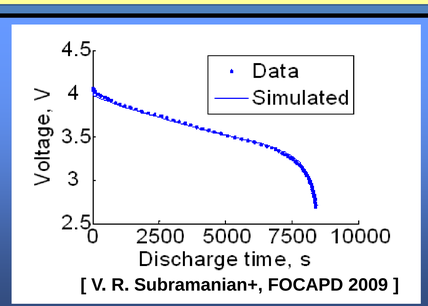
Bayesian Estimation Results



Bayesian based parameter estimation



Bayesian estimation-Capacity fade analysis



Bayesian estimation-Capacity fade prediction

Acknowledgments

The authors are thankful for the partial financial support of this work by the National Science Foundation (CBET 0828002 and 0828123) and the United States government.