Identification of Electrode Aging Mechanisms Using Mechanical Particle Model (MPM) for Lithium-Ion Batteries

Thrust Area 4: Advanced and Hybrid Powertrains

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Problem Statement

Battery degradation is currently estimated based on capacity fade and resistance increase. We aim to estimate the degradation mechanisms by identifying each electrode capacity and stoichiometry.

Approach: Fusion of voltage and strain measurements via an electrochemical and electromechanical model of the battery electrodes.

Electrochemical (Voltage):

The stoichiometric operating window of electrodes:

- \( x \): moles of Lithium in anode: \( Li_xC_6 \) with initial \( x \in (x_0, x_{100}) \)
- \( y \): moles of Lithium in cathode: \( Li_yFePO_4 \) with initial \( y \in (y_0, y_{100}) \)

The capacity of individual electrodes:
- Anode: \( C_{a0} (Ah) \)
- Cathode: \( C_{c0} (Ah) \)

At open circuit the terminal voltage can be used to identify each electrode capacity and operating stoichiometry:

Analysis using voltage-only measurements shows weak observability:

\[
S = \begin{bmatrix}
\alpha_1 & 0 & \beta_1 & \alpha_2 & 0 & \beta_2 & \ldots & \alpha_n & 0 & \beta_n \\
\alpha_1 & \beta_1 & \alpha_2 & 0 & \beta_2 & \ldots & \alpha_n & 0 & \beta_n & \ldots
\end{bmatrix}
\]

where the slopes of the overpotentials are \( \alpha_i = \frac{\partial \psi_0}{\partial x_i}, \beta_i = \frac{\partial \psi_0}{\partial y_i} \)

As an example, the error in estimating \( y_0 \) starting with a 5% initial error shows a non-observable problem when the voltage measurements are taken in the mid-SOC range, where the overpotentials are flat.

Electromechanical (Strain):

Material expansion due to Li-ion intercalation has a distinct behavior associated with material phase changes:

- Cathode active material is made of metal oxides that expands linearly with Li-ion concentration \( \epsilon_s(x) \)
- Anode active material is made of graphite that expands piece-wise linearly with Li-ion concentration based on its phase changes \( \epsilon_s(x) \)

Assuming rigidity for other components the total strain can be written as:

\[
\epsilon_T = \epsilon_p + \epsilon_s = \epsilon_p + \epsilon_n(x) + \epsilon_n(x) + C_n \epsilon_n^2 + \epsilon_n^2
\]

where \( \epsilon_p, \epsilon_n, \epsilon_n^2 \) and \( \epsilon_n^2 \) are the thickness of anode, cathode, and cell at the discharge state, respectively.

Diffusion Driven Dynamics:

A Mechanical Particle Model (MPM) to model voltage and strain concurrently. Based on Single Particle Model (SPM) with a moving boundary.

Analysis using voltage-only measurements shows weak observability:

\[
S = \begin{bmatrix}
\alpha_1 & 0 & \beta_1 & \alpha_2 & 0 & \beta_2 & \ldots & \alpha_n & 0 & \beta_n \\
\alpha_1 & \beta_1 & \alpha_2 & 0 & \beta_2 & \ldots & \alpha_n & 0 & \beta_n & \ldots
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[1] http://epg.eng.ox.ac.uk/content/degradation