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Dataset Title: Data of Degradation and Expansion of Lithium-Ion Batteries with Silicon/Graphite Anodes

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Key Points:

1. Increased pretension forces (34–172 kPa) reduced resistance and expansion with limited impact on capacity fade.
2. Cycling at 45°C extended battery life but caused more early-life resistance growth.
3. Irreversible expansion closely aligned with resistance patterns at 25°C and 0°C.
4. ICA/DVA identified LLI pre-knee and a combined effect of LLI, LAM-Anode (particularly in Si) post-knee.
5. EIS revealed SEI resistance as the dominant contributor to kinetic degradation.

Research Overview:

Lithium-ion batteries with silicon/graphite (Si/Gr) anodes achieve higher energy densities but face challenges such as rapid capacity fade, resistance growth, and complex expansion behavior under various cycling conditions. This study systematically addresses these challenges through a comprehensive test matrix to investigate the effects of pressure, temperature, state-of-charge (SoC) windows, and charge rates (C-rates) on the evolution of expansion, resistance, and capacity behavior over the lifetime of the battery. Increasing the applied pressure between 34 and 172 kPa reduced both reversible and irreversible expansion per cycle, as well as resistance growth over time, without significantly impacting capacity fade. Electrochemical Impedance Spectroscopy (EIS) confirmed that increased pressure lowered initial solution resistance and mitigated the growth of the solution and solid electrolyte interphase (SEI) resistance. Elevated temperature (45°C) extended battery cycle life despite an initial increase in resistance. Under these conditions, the lifetime impedance increase was dominated by SEI resistance. Consistent with prior studies, operating in a narrow SoC window at high SoC minimized capacity loss. Additionally, charge rates up to 2C had a limited effect on the overall degradation trends. Incremental capacity analysis (ICA) and differential voltage analysis (DVA) identified lithium inventory loss (LLI) as the primary driver of pre-knee degradation, whereas post-knee degradation resulted from a combination of LLI and anode-active material loss, particularly silicon. The deeper understanding of degradation mechanisms in batteries with Si/Gr anodes provided by this work enables the optimal packaging design and selection of operating conditions for the battery management system to extend battery cycle life.

Methodology:

1. An extensive test matrix was implemented to examine the cell-level degradation behavior on batteries with Si/Gr anodes.
2. Capacity, resistance, reversible expansion, and irreversible expansion were analyzed on 16 groups of cells. The voltage and current data were collected by the NEWARE (Model NO: MHW-200-110V) and ARBIN battery cyclers. Expansion data were collected by a specially designed fixture in our lab. EIS data were collected by BIOLOGIC BCS-815.
3. ICA and DVA were implemented to identify thermodynamic degradation mechanisms.
4. An equivalent circuit model was utilized to estimate parameters from measured EIS to identify the kinetic degradation mechanism.

Files contained:

1. [Processed Data] Figure 2

This dataset contains the a) capacity, b) resistance, c) reversible expansion, and d) irreversible expansion of cells in selected groups. The column heads include EFC(n): equivalent full cycles; Capacity (Ah), Resistance (Ohm), RevExpansion (reversible expansion, um), and IrrExpansion (irreversible expansion, um). NaN is used for padding purposes. Please remove by using data.

Figure2\_a.csv

Figure2\_b.csv

Figure2\_c.csv

Figure2\_d.csv

1. [Processed Data] Figure 3

This dataset contains the a) capacity, b) resistance, c) reversible expansion, and d) irreversible expansion of cells in selected groups. The column heads include EFC(n): equivalent full cycles; Capacity (Ah), Resistance (Ohm), RevExpansion (reversible expansion, um), and IrrExpansion (irreversible expansion, um). NaN is used for padding purposes. Please remove by using data.

Figure3\_a.csv

Figure3\_b.csv

Figure3\_c.csv

Figure3\_d.csv

1. [RAW & Processed Data] Figure 4

This dataset contains raw C/20 charging data (Q (Ah): charged capacity, V(volt): voltage) and processed dQdV an dVdQ data.

RPT\_EOL\_25degC.csv

RPT\_PreKnee\_25degC.csv

RPT\_BOL\_25degC.csv

RPT\_EOL\_45degC.csv

RPT\_PreKnee\_45degC.csv

RPT\_BOL\_45degC.csv

1. [RAW & Processed Data] Figure 5 & 6
	1. Raw EIS data for different groups at the begin-of-life (BOL) and end-of-life (EOL) correspond to Figure 5 a) and d) and Figure 6 a) and d).

This data contains EIS measured at different SoCs, including the time domain data (adjusting SoC - time/s; Ewe/V; I/mA) and frequency domain data (EIS measurements - freq/Hz; |Z|/Ohm; Phase(Z)/deg).

Group1\_EIS\_BOL.csv

Group1\_EIS\_EOL.csv

Group2\_EIS\_BOL.csv

Group2\_EIS\_EOL.csv

Group3\_EIS\_BOL.csv

Group3\_EIS\_EOL.csv

Group4\_EIS\_BOL.csv

Group4\_EIS\_EOL.csv

Group5\_EIS\_BOL.csv

Group5\_EIS\_EOL.csv

* 1. Estimated equivalent circuit model parameters with the EIS raw data correspond to Figure 5 b) c) e) f) and Figure 6 b) c) e) f).

The columns include Rs: solution resistance; R1: SEI resistance (Rsei); R2: Charge transfer resistance (Rct); tau1: time constant for RseiC1; tau2: time constant for RctC2; Aw: Walburg coefficient. RMSE%: Percentage RMSE.

Estimated\_EIS\_Paras\_Group1\_EIS\_BOL.csv

Estimated\_EIS\_Paras\_Group1\_EIS\_EOL.csv

Estimated\_EIS\_Paras\_Group2\_EIS\_BOL.csv

Estimated\_EIS\_Paras\_Group2\_EIS\_EOL.csv

Estimated\_EIS\_Paras\_Group3\_EIS\_BOL.csv

Estimated\_EIS\_Paras\_Group3\_EIS\_EOL.csv

Estimated\_EIS\_Paras\_Group4\_EIS\_BOL.csv

Estimated\_EIS\_Paras\_Group4\_EIS\_EOL.csv

Estimated\_EIS\_Paras\_Group5\_EIS\_BOL.csv

Estimated\_EIS\_Paras\_Group5\_EIS\_EOL.csv

1. [Algorithm] L-Aw-Rs-RseiC1-RctC2 equivalent circuit model parameter fitting algorithm. This algorithm estimates the model parameters and saves the parameters in the format of the files in 4.b.

EIS\_Model\_and\_Parameter\_Fitting.m

LICNESE

Naming Convention:

1. Figure **m** **n**): Figure **m** subplot **n** in manuscript.
2. RPT\_BOL(EOL)\_**xx**degC: Begin-og-Life(End-of-life) RPT data for cells cycled at **xx** degreeC.
3. Group**m**\_EIS\_BOL(EOL): Begin-og-Life(End-of-life) EIS raw data for group **m**.
4. Estimated\_EIS\_Paras\_Group**m**\_EIS\_BOL: Estimated EIS equivalent circuit model parameters for BOL EIS data of group **m**.

Abbreviations:

BOL: begin-of-life

EOL: end-of-life

RPT: reference performance test

EIS: electrochemical impafance spectroscopy

ICA: incremental capacity analysis

DVA: differential voltage analysis.

NaN: not a number, used for data padding. Remove when processing data.

Use and Access:

This data set is made available under a **Attribution 4.0 International (CC BY 4.0)**.

To Cite Data:

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Impact of Pretension, Temperature, C-rate and State-of-Charge Window