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Supplementary Materials

Capacity Degradation and Aging Mechanisms Evolution of Lithium-Ion Batteries under Different Operation Conditions

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Abstract: Since lithium-ion batteries are rarely utilized in their full state-of-charge (SOC) range (0%–100%) in practice, understanding their performance degradation with different SOC swing ranges is critical for optimizing battery usage. We modeled battery aging under different depths of discharge (DODs), SOC swing ranges and temperatures by coupling four aging mechanisms, including the solid–electrolyte interface (SEI) layer growth, lithium (li) plating, particle cracking, and loss of active material (LAM) with a P2D model. Additionally, the mechanisms causing the accelerated capacity to drop near the battery's end of life (EOL) were investigated systematically. The results indicate that when the battery operated with a high SOC range, its capacity was more prone to accelerated degradation near the EOL. Among the four degradation mechanisms, li plating was mainly sensitive to the operation temperature and SOC swing ranges, while SEI growth was mainly sensitive to temperature. Furthermore, there is an inhibitory interaction between li plating and SEI growth, as well as positive feedback between LAM and particle cracking during battery aging. Additionally, we discovered that the extremely low local porosity around the anode separator could cause the 'knee point' of capacity degradation.

Keywords: battery aging modeling; aging mechanisms evolution; capacity degradation; aging mechanisms interaction

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Table S1. Default degradation parameters.

Symbol	Definition	Negative electrode (−)	Positive electrode (+)
A	Total planar electrode area, m^2	0.1027	0.1027
$a_\pm$	Surface area to volume ratio, m^{-1}	3.84×10^5	3.82×10^5
$c_{m\pm}$	Maximum Li+ concentration, $mol \cdot m^{-3}$	33133	63104
$c_{0\pm}$	Initial Li+ concentration, mol m^{-3}	29866	17038
$D_\pm$	Li+ diffusion coefficient at 25 C, $m^2 \cdot s^{-1}$	3.3×10^{-14}	4×10^{-15}
$E_{D\pm}$	Activation energy for Li+ diffusion, $J \cdot mol^{-1}$	30300	25000
$E_{k\pm}$	Activation energy for rate constant, $J \cdot mol^{-1}$	35000	17800
$k_\pm$	(De)intercalation rate constant at 25 C, $m \cdot s^{-1}$	2.12×10^{-10}	1.12×10^{-9}
$r_\pm$	Electrode particle radius, m	5.86×10^{-6}	5.22×10^{-6}
$\delta_\pm$	Electrode thickness, m	8.52×10^{-5}	7.56×10^{-5}
ε_e	Electrode porosity	0.25	0.335
ε_a	Active material volume fraction	0.75	0.665
$\sigma_\pm$	Electrode conductivity, $S \cdot m^{-1}$	215	0.18
$c_{sol,0}$	Bulk solvent concentration, $mol \cdot m^{-3}$	2636	-
$\overline{V}_{SEI}$	SEI partial molar volume, $m^3 \cdot mol^{-1}$	9.585×10^{-5}	-
ρ_{SEI}	SEI resistivity, $\Omega \cdot m$	2×10^5	-
$L_{SEI,0}$	Initial SEI thickness, m	5×10^{-9}	-
E_{sol}	Solvent diffusion activation energy, $J \cdot mol^{-1}$	37000	-
$\alpha_{a,Li}$	Anodic transfer coefficient for Li stripping	0.35	-
$\alpha_{c,Li}$	Cathodic transfer coefficient for Li plating	0.65	-
E	Young’s modulus [Pa]	1.5×10^{10}	3.75×10^{11}
υ	Poisson’s ratio	0.3	0.2
Ω	Partial molar volume [m^3/mol]	3.1×10^{-6}	1.25×10^{-5}
$l_{cr,0}$	Initial crack length [m]	2×10^{-5}	2×10^{-5}
w_{cr}	Initial crack width [m]	1.5×10^{-5}	1.5×10^{-5}
ρ_{cr}	Number of cracks per unit area [m^{-2}]	3.18×10^{15}	3.18×10^{15}
b_{cr}	Stress intensity factor correction	1.12	1.12
m_{cr}	Paris’ law exponential term	2.2	2.2
σ_c	Critical stress for particle fracture [Pa]	6×10^7	3.75×10^8
m_2	Loss of active material exponential term	2	2

Table S2. Test matrix for lifetime cycling.

Symmetric	T = 5 °C	T = 15 °C	T = 25 °C	T = 35 °C	T = 45 °C
DOD = 30%			65%–95% 、 55%–85% 、 45%–75% 、 35%–65% 、 25%–55%、15%–45%、5%–35%		
DOD = 40%			55%–95% 、 45%–85% 、 35%–75% 、 25%–65% 、 15%–55%、5%–45%		

DOD 50%	=	45%–95%、35%–85%、25%–75%、15%–65%、5%–55%	45%–95%、35%–85%、25%–75%、15%–65%、5%–55%	45%–95%、35%–85%、25%–75%、15%–65%、5%–55%	45%–95%、35%–85%、25%–75%、15%–65%、5%–55%	45%–95%、35%–85%、25%–75%、15%–65%、5%–55%
DOD 60%	=			35%–95%、25%–85%、15%–75%、5%–65%		