



Severin Lukas Hahn (Autor)
**Lifetime prediction on lithium-ion battery cell and
system level**

Herausgeber: Prof. Dr. Kai Peter Birke

ENERGIE & NACHHALTIGKEIT
Elektromobilität & Batterietechnologie

Severin Lukas Hahn

**Lifetime prediction on
lithium-ion battery cell
and system level**

Elektrische
Energiespeichersysteme



Nachhaltige
CO₂-Kreisläufe



Elektromobilität &
Batterietechnologie



Cuvillier Verlag Göttingen
Internationaler wissenschaftlicher Fachverlag

<https://cuvillier.de/de/shop/publications/8677>

Copyright:
Cuvillier Verlag, Inhaberin Annette Jentsch-Cuvillier, Nonnenstieg 8, 37075 Göttingen,
Germany
Telefon: +49 (0)551 54724-0, E-Mail: info@cuvillier.de, Website: <https://cuvillier.de>

Table of Contents

Table of Contents	i
List of abbreviations	viii
German Abstract: Zusammenfassung	x
Abstract	xi
Chapter 1 Introduction	1
1.1 Motivation and goals of the thesis.....	2
1.2 Contributions and structure of the thesis	6
Chapter 2 Theoretical Foundation	11
2.1 Lithium-ion history and fundamentals	11
2.2 Cell construction and format considerations.....	15
2.3 Competing cell formats	17
2.4 Changing electrochemical environments	18
2.4.1 Operation principles.....	21
2.4.2 Combined electrode reactions and self-discharge	24
2.4.3 Anodic side reactions and solid electrolyte interphase	25
2.4.4 Lithium loss due to plating	27
2.4.5 Lithium loss due to cyclic SEI growth	29
2.4.6 Cathodic side reactions and solid permeable interphase	29
2.4.7 Transition metal dissolution	31
2.4.8 Loss of active material	32

2.4.9 Overhang effect.....	34
2.5 Non-destructive aging analysis	35
2.5.1 Loss of anode active material	37
2.5.2 Loss of active lithium.....	38
2.5.3 Loss of cathode active material.....	39
2.5.4 Principle of limitation	39
2.5.5 Inhomogeneity.....	40
2.5.6 Quantitative analysis by fitting	41
2.5.7 Computer tomography.....	42
2.6 Post-mortem analysis	42
2.6.1 Gas measurement.....	42
2.6.2 Disassembly	43
2.6.3 Electrode investigation	43
2.6.4 Three electrode tests	43
Chapter 3 Experimental methods.....	45
3.1 Investigated cells	45
3.2 Cycling procedures	46
3.3 Reference parameter tests during aging	46
3.4 Cyclic aging experiments.....	47
3.5 Calendar aging experiments	48
3.6 Battery scale testing	48
3.7 Electrode harvesting.....	49
3.8 Homogeneity measurements.....	50
3.9 Gas measurements	51

3.10 Half and full-cell measurements	51
3.11 Inductive coupled plasma – optical emission spectroscopy.....	52
3.12 Scanning electrode microscopy.....	53
Chapter 4 Study on Calendaric Aging Models	55
4.1 Chapter summary	55
4.2 Introduction and insights from literature.....	55
4.2.1 Structure and properties of SEI	57
4.2.2 Modeling approached for SEI growth induced aging.....	57
4.3 Extended calendar aging model development	63
4.4 Quantitative validation technique	69
4.5 Calendar aging matrix.....	70
4.6 Accelerated aging matrix results	71
4.7 Aging models	76
4.8 Validation results and detection of overfitting models	76
4.9 Extended model results.....	80
4.10 Quantitative comparison of aging models	80
4.11 Discussion	82
Chapter 5 Actively-controlled pneumatic press	85
5.1 Chapter summary	85
5.2 Introduction and insights from literature.....	86
5.3 Cell press design	90
5.3.1 Mechanics	90
5.3.2 Pneumatics	91
5.3.3 Cooling.....	92

5.3.4 Control system	93
5.4 Cell press operation modes	94
5.4.1 Touch procedure	94
5.4.2 Stiffness measurement	94
5.4.3 Constant force control.....	95
5.4.4 Module stiffness simulation.....	95
5.5 Validation results.....	97
5.6 Homogeneity validation	97
5.6.1 Setup stiffness.....	100
5.6.2 Cell operation at constant force.....	101
5.6.3 Cell operation in module stiffness simulation	103
5.7 Requirement assessment.....	109
5.7.1 Press validation	109
5.7.2 Constant force control.....	112
5.7.3 Module stiffness simulation.....	113
5.8 Discussion.....	115
Chapter 6 Pressure Prediction Modeling.....	120
6.1 Chapter summary	120
6.2 Introduction and insights from literature.....	121
6.3 Investigated battery system	123
6.4 Mechanical characterizations	125
6.4.1 Cell compressive stiffness	125
6.4.2 Module spreading test	125
6.4.3 Module pressure measurement	126

6.5 Electrical characterization overview	126
6.5.1 Normative aging	127
6.6 Electromechanical characterizations	128
6.6.1 Reversible swelling.....	128
6.6.2 Plating and SEI cycles	128
6.6.3 Gas influence.....	129
6.6.4 Cell growth parametrization	129
6.6.5 Battery level validation tests	129
6.7 Post-mortem analysis	130
6.8 Review of cell growth mechanisms.....	130
6.8.1 Normative aging results	131
6.8.2 Electrode contributions to growth	132
6.8.3 Diffusion induced strain.....	133
6.8.4 Graphite exfoliation	134
6.8.5 Gas evolution	135
6.8.6 Growth of solid electrolyte interphase.....	136
6.8.7 Lithium plating	139
6.9 Model development	140
6.10 Modeling Results.....	144
6.10.1 Mechanical parametrization.....	144
6.10.2 Bracing predictions at begin of life	146
6.10.3 Pressure evolution over lifetime	149
6.11 Discussion	150
Chapter 7 Conclusion	153

7.1 Summary	153
7.2 Perspective	154
7.3 Outlook.....	155
Appendix A	157
A.1 Electrochemical series of metals	157
Appendix B	158
B.1 Calendaric commutativity for calendaric aging models	158
B.2 Derivation of the extended model based on Broussely <i>et al.</i> [31]	159
B.3 Cell parameters for calculation of Li loss by ICP-OES	161
B.4 Aging behavior of the 40 % and 100 % SOC cell at 85 % SOC	162
B.5 Comparison of voltage dependencies	163
B.6 Fitting of $t^{0.5}$ and $t+t^{0.5}$ models	163
Appendix C.....	164
C.1 PID control algorithm.....	164
C.2 Cycling protocol for cell B.....	165
C.3 Recurring control errors	166
C.4 Supplementary homogeneity measurements.....	167
Appendix D	171
D.1 Detailed concept explanations	171
D.2 Comparative Plating and SEI cycling results.....	173
D.3 Supplementary post-mortem results	173
D.4 Simulative parameters	177
D.5 Growth aging mechanisms	178
D.6 Pure elastic model results.....	180

D.7 Cell thickness distribution.....	180
References.....	181
Curriculum vitae.....	197
List of Figures	199
List of Tables.....	209
