Towards unified machine learning characterization of lithium-ion battery degradation across multiple levels: A critical review

Alan G. Li[a], Alan C. West[b], Matthias Preindl[a,*]

[a] *Department of Electrical Engineering, Columbia University in the City of New York, 500 W. 120th St., Mudd 1310, New York, NY, 10027, USA*

[b] *Department of Chemical Engineering, Columbia University in the City of New York, 500 W. 120th St., Mudd 801, New York, NY, 10027, USA*

Abstract

Lithium-ion battery (LIB) degradation is often characterized at three distinct levels: mechanisms, modes, and metrics. Recent trends in diagnostics and prognostics have been heavily influenced by machine learning (ML). This review not only provides a unique multi-level perspective on characterizing LIB degradation, but also highlights the role of ML in achieving higher accuracies with accelerated computation times. We survey the state-of-the-art in degradation research and show that existing techniques lay the foundations for a unified ML method – a single tool for characterizing degradation at multiple levels. This could inform optimal management of lithium-ion systems, thus extending lifetimes and reducing costs. We outline a framework for the hypothesized technique and identify the challenges and future trends in degradation research. It is shown that pulse-injection has high potential, and that further work is needed for in-situ diagnostics of degradation mechanisms.

Keywords: `Battery management systems, Machine Learning, Lithium batteries,`

1. Introduction

Lithium-ion batteries (LIBs) are central to the decarbonization of transport and the integration of renewable energy into the grid. Technologies like electric vehicles (EVs) and grid storage are likely to become reliant on LIBs in the coming years due to their high energy densities, efficiency, and reliability [1]. The future of battery research lies not only in next-generation chemistries such as lithium-air or solid-state [2, 3], but also better management of existing commercial cells such as $LiNi_xMn_yCo_{1-x-y}O_2$ (NMC), $LiNi_xCo_yAl_{1-x-y}O_2$ (NCA), and $LiFePO_4$ (LFP) [4, 5]. Developing advanced battery management systems (BMS) is therefore a key factor in driving down battery costs and promoting widespread adoption.

Advanced battery management is essentially synonymous with big data and machine learning (ML) [6, 7]. Though safety remains the fundamental task of the BMS, decentralized architectures using 'smart cells' could allow the BMS to actively extend cell lifetime. Achieving this goal depends on a better understanding of LIB degradation. Novel sensing technologies such as strain sensors and optical fibers have been proposed for this purpose, but the cost and computational burden of big data remain challenges [8, 9]. Meanwhile, ML techniques using traditional sensing techniques – voltage, current, and temperature – have been successful at both degradation diagnostics and prognostics [10].

Battery degradation research traditionally lies in the intersection of electrical and chemical engineering. This is because there are multiple levels of detail from which to examine LIB degradation, as shown in Figs. 1 and 2. Several studies have organized degradation in a similar fashion [12, 13, 14, 15, 16], though rarely is each level examined in detail. Often the internal mechanisms and modes of degradation are irrelevant for basic BMS functions, and only the performance metrics are needed. Yet it has been shown that deeper knowl-

*Corresponding author.
Email address: `matthias.preindl@columbia.edu` (Matthias Preindl)

Nomenclature

BMS	Battery management system
DTV	Differential thermal voltammetry
DVA	Differential voltage analysis
EIS	Electrochemical impedance spectroscopy
EV	Electric vehicle
GITT	Galvanostatic intermittent titration technique
GPR	Gaussian process regression
ICA	Incremental capacity analysis
LAM	Loss of active material
LFP	$LiFePO_4$
LIB	Lithium-ion battery
LLI	Loss of lithium inventory
ML	Machine learning
NCA	$LiNi_xCo_yAl_{1-x-y}O_2$
NE	Negative electrode
NMC	$LiNi_xMn_yCo_{1-x-y}O_2$
NN	Neural network
OCV	Open circuit voltage
P2D	Pseudo-2D
PBM	Physics based model
PE	Positive electrode
RNN	Recurrent neural network
RUL	Remaining useful life
SEI	Solid electrolyte interphase
SoC	State of charge
SoH	State of health
SoP	State of power
SPM	Single particle model
SVM	Support vector machine

edge of degradation due to various stress factors can yield tangible cost benefits by informing advanced control strategies [17, 18, 19, 20, 21]. Hence the need to assess degradation at multiple levels.

At the highest level, degradation is characterized using performance metrics including state of health (SoH), remaining useful life (RUL), and state of power (SoP). Metrics capture the effects of capacity and power fade and are most easily observed by the BMS. As such, they have received the most attention. State estimation is a highly-researched field with several mature techniques used to provide basic diagnostics information about LIB cells such as state of charge (SoC) in addition to SoH and SoP [22, 23]. LIB states are 'backwards-looking' metrics, however, and RUL is used for degradation prognostics [24]. Related to RUL is the colloquial 'knee-point' observed in capacity fade curves, which represents the point at which LIB degradation accelerates and renders the cell unsuitable for EV applications [25, 26].

The evolution of degradation metrics over time is explained by degradation modes. Commonly reported modes include the loss of lithium inventory (LLI), and the loss of active material (LAM) at the postive electrode (PE) or negative electrode (NE), though stoichiometric drift between the electrodes and impedance change are also mentioned [12, 27]. LLI and LAM are valuable quantities that have been shown to accurately evaluate and predict LIB voltage and degradation behavior [28].

Modes result from degradation mechanisms in internal cell components, represented in Fig. 3. Various chemical processes occur in the cell during cycling. They are strongly coupled and experience intricate positive and negative feedback loops. Both electrodes are subject to fracturing due to the volumetric strain of repeated lithiation cycles. In the graphite NE, Li ions may be deposited on the surface instead of intercalating, known as lithium plating. Solid electrolyte interphase (SEI) layers exacerbate the situation by forming on the NE and plated lithium surfaces, trapping lithium ions and impeding ion movement [27]. While the PE is less subject to SEI formation and plating, it is particularly subject to chemistry-specific structural and chemical change that reduce active material. Some mechanisms may be destructive – SEI decomposition can lead to thermal runaway and dendrite growth from plated lithium can cause internal shorting [29]. Detailed knowledge of internal mechanisms is crucial for ensuring safety and extending lifetime.

There have been few, if any, attempts to characterize degradation mechanisms, modes, and metrics with a single technique in real-time. It has been suggested that doing so could extend battery lifetime and reduce costs [27]. Traditional

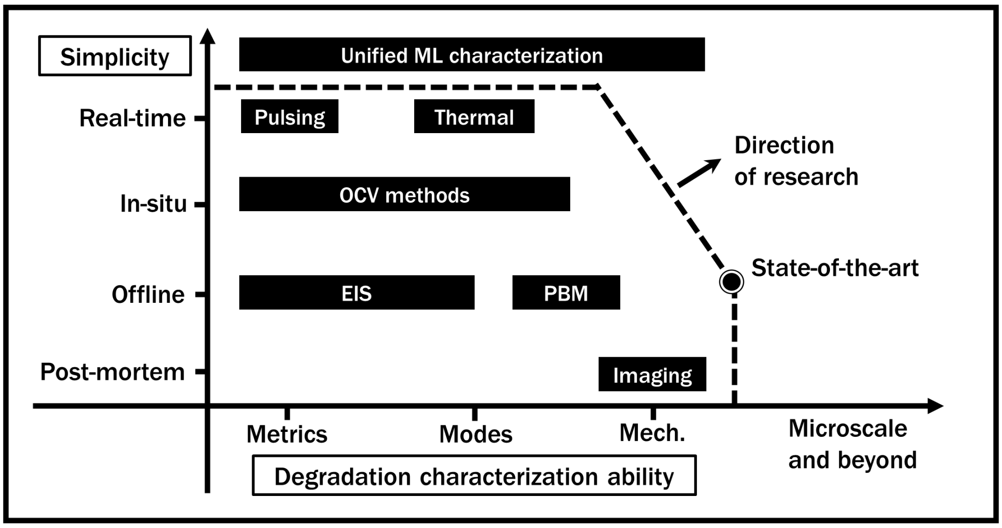

Figure 1: Simplicity and characterization ability of various diagnostics techniques for degradation at multiple levels, adapted from [11]. Research trends are moving towards a unified ML framework located beyond the current state-of-the-art.

techniques to measure degradation often rely on measurements from ex-situ controlled environments [30], or physics-based models with high computational complexity [31]. These methods are infeasible for real-time estimation. In contrast, ML methods such as neural networks (NNs), support vector machines (SVMs), or Gaussian process regression (GPR) can offer real-time estimation given sufficiently high-quality training data and selection of an appropriate input signal [32].

1.1. Contributions and Outline

To date, there has not been a critical perspective assessing ML techniques for characterizing LIB degradation stress factors, mechanisms, modes, and metrics. We offer insight into the strengths and challenges of using ML for non-invasive multi-level degradation diagnostics. The possibility for a single ML technique to uncover degradation at multiple levels – which we term 'unified characterization' – is evaluated from a wide range of studies. This review thus provides the theoretical foundations towards a unified ML-based LIB degradation diagnostics technique.

The review continues in Section 2, where key review papers on LIB degradation, machine learning for LIB systems, and their intersection are identified and their insights discussed. In Section 3, state-of-the-art techniques for degradation characterization at all levels are discussed, with particular attention to ML-based methods. In Section 4, a framework for unified ML degradation characterization is proposed and assessed. The review is then concluded in Section 5.

2. Key Reviews on Degradation

Reviews on degradation research are ostensibly quite disparate. In one category are reviews that focus on degradation processes and their link to stress factors or cell chemistry. Others focus on diagnosing degradation mechanisms and modes. Still others focus on SoH and RUL estimation for use in applications such as EVs or grid services. These apparently distinct types of reviews are, however, unified by their praise of ML techniques [33]. This section aims to examine all types of reviews and point towards key sources of information, summarized in Table 1.

A multi-level framework for understanding LIB degradation was first developed no later than 2005 by Vetter et al. [12], in which the mechanisms, modes, metrics, and stress factors are listed and described. A decade later, similar processes were identified in [34] but with greater focus on destructive failures such as short-circuits and thermal runaway. In [13], better characterization of degradation mechanisms is explicitly tied to improved performance and cycle life. The effects of temperature on degradation, such as low temperature lithium plating, are summarized in [35]. Multi-level degradation from the material level to system level is

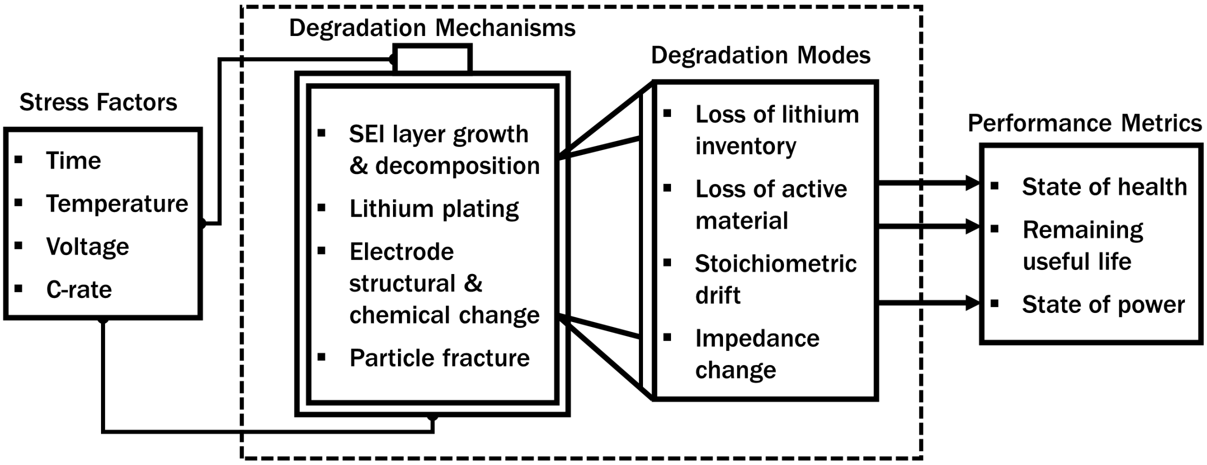

Figure 2: Summary of LIB cell stress factors and the resultant internal degradation mechanisms and modes leading to observable performance metrics.

discussed in [15], with consideration of variations in cell design and fabrication. In [36], a wide range of chemistry- and electrode-specific mechanisms are discussed in detail. A consumer's perspective is presented in [37], providing helpful recommendations based on academic results.

Diagnostics techniques for degradation mechanisms and modes are well-documented. Traditionally imaging and chemical analysis techniques such as microscopy and spectroscopy are used to experimentally verify degradation mechanisms [27, 30], but cannot be performed in a BMS. Model-based methods for mechanisms have potential for real-time use, but still have high complexity. Models for SEI formation, crack growth, and LAM are reviewed in [31]. Lithium-plating is comprehensively reviewed in [38], including imaging and model-based methods, with applications in fast charging. A general review of LIB system faults, including degradation mechanisms, is given in [29]. Diagnostics for degradation modes are reviewed in [39, 28, 40], where non-invasive techniques are discussed in detail, including electrochemical impedance spectroscopy (EIS), incremental capacity analysis (ICA), differential voltage analysis (DVA), galvanostatic intermittent titration (GITT), and pulsing. These characterization methods may also provide qualitative information on underlying mechanisms.

Reviews focusing on SoH and RUL estimation are perhaps the most mature and share similar methodologies. Some focus only on SoH [41, 42], others on RUL prognostics [24], while [16] and [43] examine both. There is general consensus that ML combined with novel sensing techniques, in contrast with model-based estimation, can offer superior accuracy and speed [16, 23, 41, 44], and indeed several studies focus entirely on ML methods [10, 32].

Berecibar et al. [41] predicted in 2016 that ML-based detection of degradation would dominate in the future. More recently, [32] and [33] concisely summarize the uses of ML as: (1) Accurate characterization of degradation across multiple levels, (2) Simulation of high-quality datasets, and (3) Accelerated computations. Data generation and accelerated computations for battery design are reviewed in [45, 46, 47], but multi-level degradation characterization is only partially addressed – a gap this review aims to fill.

3. State-of-the-Art in Degradation Research

Recent advances in ML-based degradation characterization are reviewed here with respect to stress factors, mechanisms, modes, and metrics. We do not provide a thorough technical description, rather, we aim to evaluate the benefits, challenges, and wider potential of using ML compared to traditional methods.

3.1. Stress factors

LIB systems are sensitive to extremes - high and low temperatures, high and low voltages or SoC, and high C-rates. During use, stress factors can often be mitigated by the BMS through thermal management, cycling protocols, and charging limits

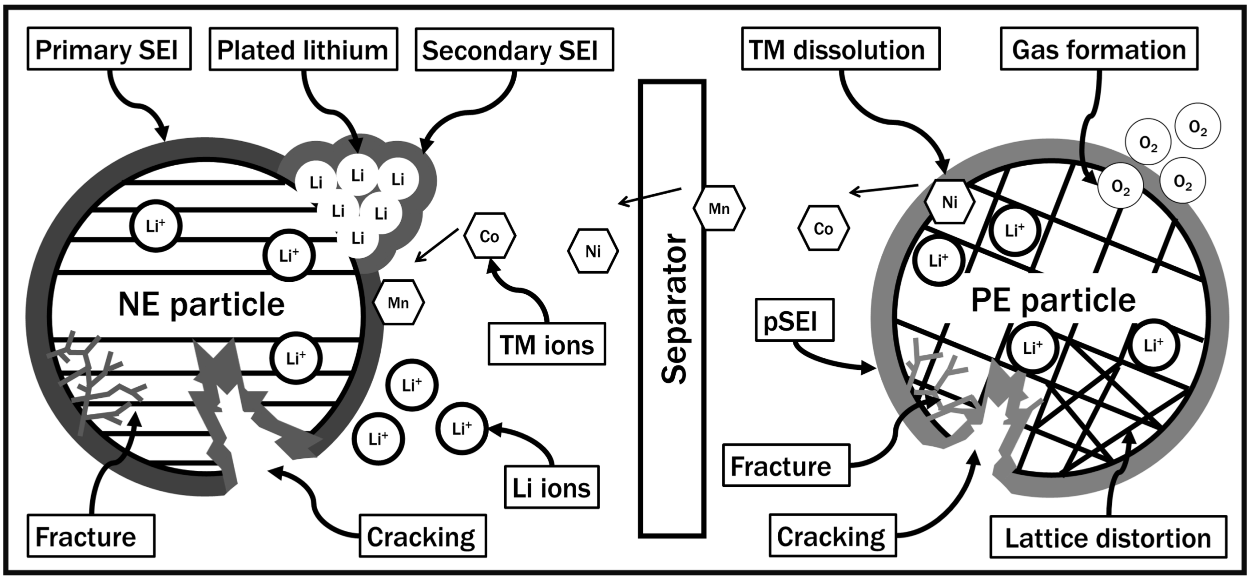

Figure 3: Exaggerated diagram of degradation mechanisms at the negative electrode (NE) and postive electrode (PE) within LIB cells, representing solid electrolyte interphase (SEI) formation, lithium plating, transition metal (TM) dissolution, gas formation, lattice distortion, and particle fracture/cracking.

[8]. Precise knowledge and quantification of the effects of certain stresses on battery lifetime remains a challenge. Knowing the most significant stressor could inform more targeted BMS strategies to extend lifetime [26, 48, 49, 50, 51, 52].

One approach to stressor analysis requires the design of specific cycling protocols. Long-term cycling is performed in [49] for commercial NCA cells subjected to various regimes of cycling and rest – cyclic and calendar aging – at a range of C-rates. It is experimentally confirmed that high C-rates induce path dependence. From EIS, ICA, and DVA, the sequence of the cycling and rest regimes is shown to alter capacity fade by inciting certain degradation mechanisms. Large datasets are also used in [48] to examine the effects of a wide range of temperatures, SoC ranges, and C-rates on NMC, LFP, and NCA cells. Degradation is shown to be chemistry-dependent: stress factors do not affect different cells in the same way.

Feature selection, a form of unsupervised ML, can offer automated insight into the most important stressors without specific cycling protocols. This may reinforce existing knowledge, as shown in [26], where various cycling features are ranked for their ability to predict RUL. Feature coupling can lead to misleading conclusions if not interpreted correctly – mid-range SoC cycling was the most important feature, which may appear to contradict prior results, but [26] explain that this was highly correlated with cycling at low and high SoC levels, which is known to exacerbate degradation. Stressor ranking is also performed in [50] with random forests to examine the effects of temperature, charging and discharging rates, cut-off current, and SoC levels. Consideration of coupling between stressors shows that high temperature and discharge currents had the highest importance. Features are also selected in [51] and [52] for SoH estimation using ML, where up to 30 voltage, current, and temperature features extracted from cycling data or charging curves are considered. Analysis of the most important features could reduce the large data processing requirements inherent in these techniques.

3.2. Mechanisms

Diagnosing degradation mechanisms is a complex task, requiring either destructive post-mortem analysis or non-invasive diagnostics. Both methods are used in [53] to examine the effects of extreme low temperature and SoC. Lithium plating is verified as the primary mechanism using qualitative observations. Practical applications, however, would require a more detailed quantitative understanding of mechanisms. This can be addressed with ML methods, as proposed in [54, 55, 56, 57].

Post-mortem analyses are performed in [54] and [55]. X-ray spectroscopy and tomography were

Table 1: Summary of key review papers related to LIB degradation (deg.) and machine learning. 'Multi-level' papers consider stress factors, mechanisms (mech.), modes, and metrics.

Review paper	Levels	Focus	Key insight
X. Chen et al., 2021 [33]	Multi-level	General	ML can be applied to LIB for multi-level applications
Woody et al., 2020 [37]	Multi-level	Extending lifetime	Deg.-informed changes in consumer behavior can extend LIB life
Han et al., 2019 [15]	Multi-level	General	Cell design and fabrication affect deg.
Kabir et al., 2017 [13]	Multi-level	General	Better degradation diagnostics improves performance and cycle life
Hendricks et al., 2015 [34]	Multi-level	General	Destructive failures linked to deg. mech.
Vetter et al., 2005 [12]	Multi-level	General	First to create framework for multi-level deg.
Edge et al., 2021 [27]	Mech.	General	A unified deg. mech model capturing coupling and feedback would be a major achievement
X. Lin et al., 2021 [38]	Mech.	Lithium plating	Pulse charging may reduce Li plating
X. Hu et al., 2020 [29]	Mech.	Fault diagnosis	Fast and accurate sensing can improve LIB systems
Pender et al., 2020 [36]	Mech.	Electrode chemistry effects	Different mech. occur in different types of electrodes
Happuarachchi et al., 2018 [30]	Mech.	Anode diagnostics	In-situ techniques are not easily applied to commercial cells
Rodrigues et al., 2017 [35]	Mech.	Temperature effects	Temperature plays a large role on deg. mech.
Reniers et al., 2019 [31]	Mech., modes	Deg. models	Multiple deg. models should be used for higher accuracy
Xiong et al., 2020 [39]	Modes	Diagnostics	Advanced ECM can yield real-time diagnostics
Pastor-Fernandez et al., 2019 [28]	Modes	Diagnostics	Pseudo-OCV and IC-DV tests are the most promising
Barai et al., 2019 [40]	Modes, metrics	Diagnostics	Pulse injection and EIS are a good combination
Berecibar et al., 2016 [41]	Modes, metrics	General	ML can characterize degradation at multiple length levels
Sui et al., 2021 [10]	Metrics	SoH estimation	Non-probabilistic ML techniques are highly promising
Ng et al., 2020 [32]	Metrics	SoH estimation	ML can accurately degradation across multiple length levels
X. Hu et al., 2020 [24]	Metrics	RUL estimation	Model-based techniques can yield RUL and deg. modes without time-consuming aging tests
Wu et al., 2020 [44]	Metrics	Data-driven diagnostics	Cloud-based 'digital twins' will use big data and ML for multi-level diagnostics
X. Hu et al., 2019 [23]	Metrics	General	Big data and ML will be used for multi-level diagnostics
Y. Li et al., 2019 [16]	Metrics	Data-driven diagnostics	Differential analysis with ML is promising for multi-level deg. analysis
Lipu et al., 2018 [43]	Metrics	General	Data-driven diagnostics is more robust than model-based but requires big data
Xiong et al., 2018 [42]	Metrics	SoH estimation	Big data and the cloud will be used to model performance

combined with NN-based classification in [54] to quantitatively observe and analyze NMC cathode particles. Several microscale parameters such as particle size, detachment, and conductivity are directly measured, and their behavior evaluated under cycling at different C-rates. Using ML is shown to greatly aid data segmentation and computation. Similar methods are used in [55] also for NMC cathode particles, focusing on how particle cracking due to the strain of repeated lithiation cycles can affect Li ion transport and reaction kinetics. X-ray spectroscopy and NN-classification reveal the regions and degree of strain. Both [54] and [55] offer a valuable ML framework for obtaining a highly-accurate and detailed understanding of degradation mechanisms.

Alternatives to post-mortem analysis are presented in [56] and [57] with a focus on SEI formation and lithium plating. Non-invasive techniques such as EIS, ICA, and GITT can track the effects of the SEI, but ML could be used to obtain more detailed information about SEI structure by accelerating physics-based models (PBMs). A 'generative deep learning' framework proposed by [56] thus uses large amounts of data about SEI formation to predict SEI behavior in arbitrary scenarios; it is conceivable this could be extended to other degradation mechanisms. A more focused approach could focus on electrochemical signatures left by the mechanisms. By fabricating two types of cells with pre-determined susceptibility to SEI or plating, [57] confirm that the mechanisms are easily distinguishable using non-invasive diagnostics. This is promising for ML methods that can analyze more subtle signatures in commercial cells.

3.3. Modes

Degradation modes have attracted attention for their ease of observability compared to mechanisms, and the widespread use of half-cell open-circuit voltage (OCV) models that parametrize LLI and LAM. This can largely be attributed to Dubarry et al., [58, 59, 60, 61], who developed the 'Alawa' toolbox for simulating degradation modes. Their work has been experimentally validated by [14] and extended by [62, 63, 64, 65]. Much of the conclusions are drawn from ICA and DVA curves, whose peak widths and locations are highly sensitive to LLI and LAM. NN regression can be used to bypass parameter identification, as shown in [65, 66], which can facilitate real-time implementation.

Alternative methods to parametrize LLI and LAM are derived from PBM parameters [67, 68, 69]. A single-particle model (SPM) is used in [67] to estimate OCV and obtain modes. In [68], a pseudo-2D (P2D) model is used to track LAM, diffusivity, and the reaction constant in LIB cells. SPM, P2D, and an 'improved' SPM are compared in [69], where LLI and LAM are directly calculated from the PBM parameters. Diffusivity is an important measure of impedance change in [68] and [69] that can be linked to particle fracture and SEI formation. PBM parameters have high computational complexity that could be addressed using ML.

EIS is important diagnostics tool, but is mainly used for qualitative analysis of degradation. It is suggested in [70] that equivalent-circuit model (ECM) parameters from EIS diagnostics are highly correlated with LLI, particularly the charge-transfer and diffusion elements. The acceleration in degradation beyond the knee-point is attributed to LLI. This conflicts with Dubarry et al., 2012 [61], who describe two regimes of degradation: a linear regime where LAM 'incubates' and LLI dominates, then rapid capacity loss as LAM dominates. Still more conflicting interpretations are given in [71, 72], where ECM parameters are made directly proportional degradation modes; resistances are linked to LLI, while diffusion is linked to LAM. Since ECM parameters are inherently simplifications of internal processes in the cell, they likely capture coupled effects of LLI and LAM. While the evolution of electrochemically-defined ECM parameters can provide qualitative understanding of degradation [73], it remains unclear how accurately they can quantify degradation modes.

More recently, temperature methods have gained attention for characterizing degradation modes. It is shown in [74] that temperature differentials between the electrodes induces unique modes compared to ordinary operation; the colder electrode becomes more susceptible to degradation. Differential thermal voltammetry (DTV) concepts introduced in [75] are extended in [11], who parametrize LLI and LAM in a combined OCV-heat-transfer model. Using constant current charging, the degradation modes can be estimated. If accurate thermal measurements can be achieved in real LIB pack configurations then temperature methods could offer significant value to diagnostics.

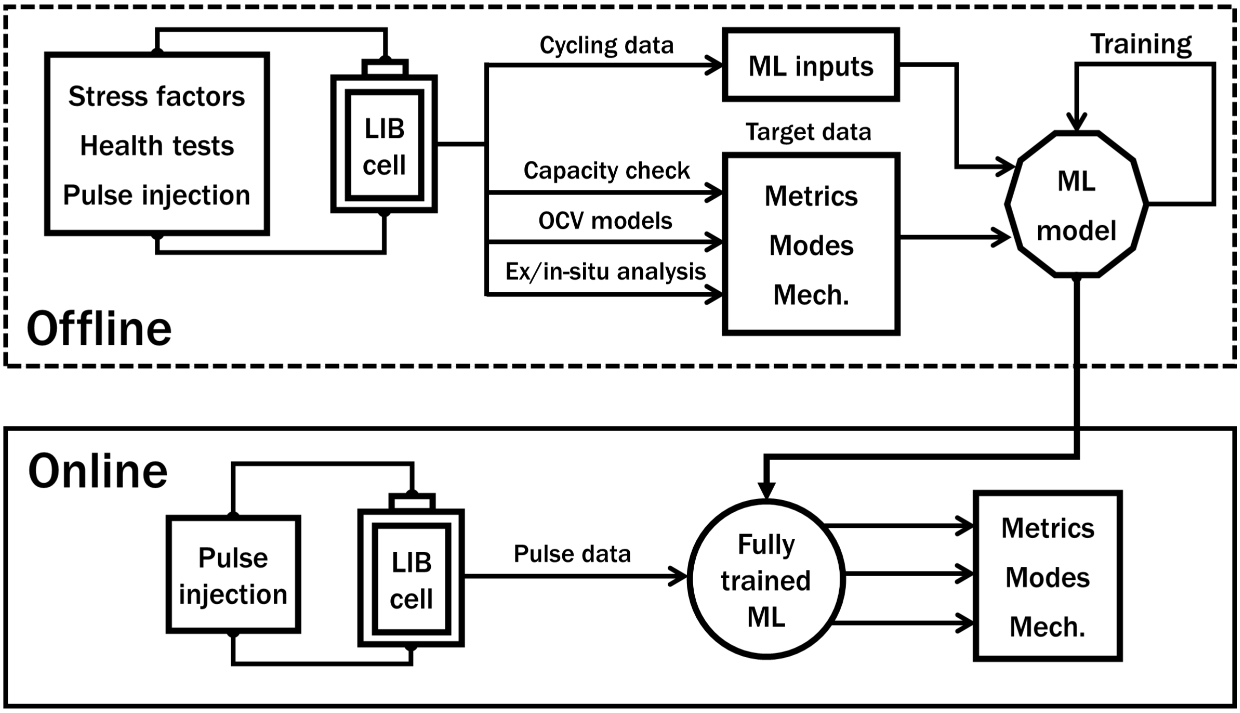

Figure 4: Proposed unified machine learning framework for diagnostics and prognostics of degradation mechanisms, modes, and metrics

3.4. Metrics

More studies use ML for SoH/RUL estimation than for any other level of degradation. Other data-driven techniques have demonstrated high performance but ML estimation has shown greater generality in addition to high accuracy [76, 77]. The various ML tools have important discrepancies, as explained in [10], but characterization frameworks are more broadly divided by the input data type. There are three types of datasets used for ML training: (1) Cycling data, such as voltage, current, and temperature, (2) Charging or discharging voltage curves, and (3) Specific diagnostics signals. Cycling data maximizes the amount of knowledge, and is often used for RUL prognostics. Charging curves can reduce the ML complexity by limiting the amount of data. Using specific diagnostics signals is a minimalist approach that can offer the highest generality with the lowest complexity. A framework combining all data types is offered in [78] using the concept of a cloud-based ‘digital twin’ that is created for each cell with beginning-of-life characterization tests and continuously updated with cycling data. Until real-time computational power can meet the demands of a cloud-based solution, careful selection of input data will still be required.

Using as much cycling data as possible has yielded highly accurate diagnostics and prognostics of LIB degradation. Earlier studies like [79, 80] estimate SoH and RUL using NNs and SVMs. Optimizing NN performance is performed with data-driven weight initialization [81] or adaptive dropout [82]. Other studies use GPR [83, 84, 85, 86], and it is shown in [87] that a similar concept can be used to create a high-fidelity OCV model for SoH prediction. Manual feature selection and processing in these studies is a contribution perhaps equally as valuable as the estimation framework itself. Automated feature selection, as noted in Subsection 3.1, is used to create a ‘pipeline’ from cycling data collection to SoH/RUL prediction using a variety of ML methods for the same purpose [26, 52, 88, 89, 90]. Early RUL prediction has become a popular subject, driven in part due to the success of Severson et al. [91] in predicting LIB lifetime primarily from the constant-discharge voltage curves of the first 100 cycles. This is improved in [92] with evaluation of a wide range of ML techniques for prediction and feature selection to achieve higher accuracy.

Limiting the amount of input data for obtaining metrics can simplify model complexity and facilitate practical implementation. Charging and discharging curves are thus popular data types; information from ICA and DVA are encoded within. Early prediction of RUL is performed in [93] with the discharge voltage of one cycle input to a convolutional NN, though error increases rapidly if performed too early in the cell's lifetime. Features from conventional charging cycles are used in [94] for diagnosing SoH with GPR. Partial charging or discharging curves, meanwhile, are particularly attractive since full cycles may not be consistently available. Data from a 25% SoC window is used in [95] and [96] to predict SoH using GPR and SVM. A range of SoC windows are examined in [97] using GPR and in [98] using the 'NARX' NN, where it is shown that width of the SoC range affects accuracy more than the position. Thus partial cycling curves trade estimation accuracy for speed.

The most minimalist of ML approaches use specific diagnostics signals. No feature selection or prior history are required, simplifying diagnostics and granting greater flexibility. A vast number of EIS spectra are shown in [99] to provide highly accurate predictions of RUL using GPR. EIS cannot be performed in real-time, but estimation using a single GITT-type pulse demonstrates fast and accurate estimation using a NN [100, 101, 102]. Since a rectangle current pulse stimulates the LIB cell at a wide range of frequencies, similar to galvanostatic EIS, pulse injection could offer similarly high performance in ML diagnostics.

4. Unified Machine-Learning Characterization

Degradation characterization is gradually extracting more information in shorter time periods to improve performance and extend LIB lifetime. The 'holy grail' of perfect real-time simulation of internal degradation mechanisms remains a difficult computational task for the near future. Existing research, however, suggests that the past and future degradation mechanisms, modes, and metrics of a LIB cell could be diagnosed with machine learning in a matter of minutes. We offer a possible framework and perceived bottlenecks for achieving this goal.

4.1. Proposed framework

Our unified framework for degradation characterization is shown in Fig. 4. As in most ML frameworks, there is an offline regime requiring large amounts of training data, computational resources, and model validation. During cycling, the cell is exposed to a wide range of stress factors and pulses are applied along with conventional LIB health tests such as pseudo-OCV capacity checks. State-of-the-art techniques should be used to characterize the cell as it ages to generate the target data. After supervised training, We hypothesize that ML can then extract the same information from pulsing data, thus 'bypassing' more complex characterization performed offline. This concept has already explored as pulse-injection-aided machine learning (PIAML) [101].

Pulse injection, a generalization of hybrid pulse power characterization, is a promising source of input data [103]. From a control theory perspective, LIB systems can be completely characterized by their transient and steady state behavior. The transient response is determined by electrochemical overpotentials and analyzed with mature techniques such as EIS, GITT, and pulsing [104, 105, 106]. The steady-state response is determined by the OCV characteristic and analyzed with ICA, DVA, and DTV. It would seem that both overpotentials and OCV are equally necessary for characterizing degradation; the studies reviewed earlier suggest otherwise. Knowing the full LIB OCV characteristic is not necessary to obtain accurate degradation metrics [95, 96, 97, 98]. Furthermore, certain SoC levels seem to 'encode' more information than others [68, 99, 103], a conclusion supported by ICA graphs. Thus it is conceivable that pulse injection at a carefully-selected SoC level could encode sufficient degradation information to train a NN.

4.2. Outlook

Degradation metrics and modes are readily obtained from existing methods; there is no consensus over the most effective. Mechanisms remain the most difficult to characterize and are fittingly the most active research area. ML-assisted microscopy/spectroscopy techniques have demonstrated high potential [54, 55] in quantifying the extent of mechanisms like SEI formation, lithium plating, particle cracking, and lattice distortion. It is likely that novel sensing techniques using acoustics, strain, or optics will be developed to allow analysis to be performed in-situ, instead of post-mortem. Advances in computational power could also facilitate the implementation of high-fidelity PBMs.

Characterizing degradation with high accuracy and resolution over a cell's lifetime could open new opportunities in battery management. The effects of stress factors on degradation would be precisely understood, which would benefit applications like fast charging [20] or vehicle-to-grid applications [17].

5. Conclusion

LIB degradation research at multiple levels was reviewed with particular attention paid to the role of ML. Characterizing the stress factors, mechanisms, modes, and metrics of degradation is a complex task with most research directed towards a single aspect. We identify several similarities between the characterization methods and hypothesize that a single ML technique can be used to provide a unified understanding of LIB degradation. This framework would use pulse injection as a diagnostics signal, allowing ML to quickly bypass traditional time-consuming characterization techniques.

Further work is needed to quantify the degradation mechanisms in LIB cells using non-destructive techniques. Novel sensing technologies are already being proposed and the continued increase in computation power could facilitate the implementation of high-fidelity PBMs in the cloud. Battery degradation is a crucial factor in determining the technical and economic feasibility of LIB systems and will remain an active research field for the years to come.

6. Acknowledgements

This research was undertaken, in part, through funding from the Columbia University Data Science Institute (DSI) Seed Fund Program.

References

[1] FCAB, Federal Consortium for Advanced Batteries (2021).
[2] T. Kim, W. Song, D. Son, L. K. Ono, Y. Qi, J. Mater. Chem. A 7 (2019).
[3] A. Masias, J. Marcicki, W. A. Paxton, ACS Energy Lett. 6 (2021) 621–630.
[4] F. H. Gandoman, J. Jaguemont, S. Goutam, R. Gopalakrishman, Y. Firouz, T. Kalogiannis, N. Omar, J. V. Mierlo, Appl. Energy 251 (2019).
[5] L. Mauler, F. Duffner, W. G. Zeier, J. Leker, Energy and Environmental Sci. 14 (2021).
[6] Y. Wang, J. Tian, Z. Sun, L. Wang, R. Xu, M. Li, Z. Chen, Renew. Sustain. Energy Rev. 131 (2020).
[7] G. dos Reis, C. Strange, M. Yadav, S. Li, Energy and AI 5 (2021).
[8] H. Dai, B. Jiang, X. Hu, X. Lin, X. Wei, M. Pecht, Renew. Sustain. Energy Rev. 138 (2021).
[9] Z. Wei, J. Zhao, H. He, G. Ding, H. Cui, L. Liu, J. Power Sources 489 (2021).
[10] X. Sui, S. He, S. B. Vilsen, J. Meng, R. Teodorescu, D. Stroe, Appl. Energy 300 (2021).
[11] R. Prosser, G. Offer, Y. Patel, J. Electrochem. Soc. 168 (2021).
[12] J. Vetter, P. Novak, M. R. Wagner, C. Veit, K. C. Moller, J. O. Besenhard, M. Winter, M. Wolhfart-Mehrens, C. Vogler, A. Hammouche, J. Power Sources 147 (2005) 269–281.
[13] M. M. Kabir, D. E. Demirocak, Int. J. Energy Rersearch 41 (2017) 1963–1986.
[14] C. R. Birkl, M. R. Roberts, E. McTurk, P. G. Bruce, D. A. Howey, J. Power Sources 341 (2017) 376–386.
[15] X. Han, L. Lu, Y. Zheng, X. Fend, Z. Li, J. Li, M. Ouyang, eTransportation 1 (2019).
[16] Y. Li, K. Liu, A. M. Foley, A. Zulke, M. Berecibar, E. Nanini-Maury, J. V. Mierlo, H. E. Hoster, Renew. Sustain. Energy Rev. 113 (2019).
[17] B. Bibak, H. Tekiner-Mogulkoc, Renewable Energy Focus 36 (2021).
[18] J. M. Reniers, G. Mulder, D. A. Howey, J. Power Sources 487 (2021).
[19] S. Nazari, F. Borelli, A. Stefanopoulou, Proceedings of the IEEE 109 (2021).
[20] A. Tomaszsewska, Z. Chu, X. Feng, S. O'Kane, X. Liu, J. Chen, C. Ji, E. Endler, R. Li, L. Liu, Y. Li, S. Zheng, S. Vetterlein, M. Gao, J. Du, M. Parkes, M. Ouyang, M. Marinescu, G. Offer, B. Wu, eTransportation 1 (2019).
[21] J. Lindgren, P. D. Lund, J. Power Sources 328 (2016) 37–45.
[22] W. Waag, C. Fleischer, D. U. Sauer, J. Power Sources 158 (2014) 321–339.
[23] X. Hu, F. Feng, K. Liu, L. Zhang, J. Xie, B. Liu, Renew. Sustain. Energy Rev. 114 (2019).
[24] X. Hu, L. Xu, X. Lin, M. Pecht, Joule 4 (2020) 310–346.
[25] P. Fermin-Cueto, E. McTurk, M. Allerhand, E. Medina-Lopez, M. F. Anjos, J. Sylvester, G. dos Reis, Energy and AI 1 (2020).
[26] S. Greenbank, D. Howey, IEEE Trans. Ind. Inf. (2021).
[27] J. S. Edge, S. O'Kane, R. Prosser, N. D. Kirkaldy, A. N. Patel, A. Hales, A. Ghosh, W. Ai, J. Chen, J. Yang, S. Li, M. Pang, L. B. Diaz, A. Tomaszewska, M. W. Marzook, K. N. Radhakrishnan, H. Wang, Y. Patel, B. Wu, G. J. Offer, Phys. Chem. Chem. Physics 23 (2021).
[28] C. Pastor-Fernandez, T. F. Yu, W. D. Widanage, J. Marco, Renew. Sustain. Energy Rev. 109 (2019) 138–159.
[29] X. Hu, K. Zhang, K. Liu, X. Lin, S. Dey, S. Onori, IEEE Ind. Electron. Magazine (2020) 65–91.
[30] S. N. S. Hapuarachchi, Z. Sun, C. Yan, Advanced Sus-

tainable Systems 2 (2018).
[31] J. M. Reniers, G. Mulder, D. A. Howey, J. Electrochem. Soc. 166 (2019) A3189–A3200.
[32] M. Ng, J. Zhao, Q. Yan, G. J. Conduit, Z. W. Seh, Nat. Mach. Intell. 2 (2020).
[33] X. Chen, X. Liu, X. Shen, Q. Zhang, Angewandte Chemie Int. Ed. 60 (2021).
[34] C. Hendricks, N. Williard, S. Matthew, M. Pecht, J. Power Sources 297 (2015) 113–120.
[35] M. F. Rodrigues, G. Babu, H. Gullapalli, K. Kalaga, F. N. Sayed, K. Kato, J. Joyner, P. M. Ajayan, Nat. Energy 2 (2017).
[36] J. P. Pender, G. Jha, D. H. Youn, J. M. Ziegler, I. Andoni, E. J. Choi, A. Heller, B. S. Dunn, P. S. Weiss, R. M. Penner, C. B. Mullins, ACS Nano 14 (2020) 1243–1295.
[37] M. Woody, M. Arbabazadeh, G. M. Lewis, G. A. Keoleian, A. Stefanopoulou, J. Energy Storage 28 (2020).
[38] X. Lin, K. Khosravinia, X. Hu, W. Lu, Progress in Energy and Combustion Sci. 87 (2021).
[39] R. Xiong, Y. Pan, W. Shen, H. Li, F. Sun, Renew. Sustain. Energy Rev. 131 (2020).
[40] A. Barai, K. Uddin, M. Dubarry, L. Somerville, A. McGordon, P. Jennings, I. Bloom, Progress in Energy and Combustion Sci. 72 (2019) 1–31.
[41] M. Berecibar, I. Gandiaga, I. Villarreal, N. Omar, J. V. Mierlo, P. V. den Bossche, Renew. Sustain. Energy Rev. 56 (2016) 572–587.
[42] R. Xiong, L. Li, J. Tian, J. Power Sources 405 (2018) 18–29.
[43] M. S. H. Lipu, M. A. Hannan, A. Hussain, M. M. Hoque, P. J. Ker, M. H. Saad, A. Ayob, J. Clean. Prod. 205 (2018) 115–133.
[44] B. Wu, D. Widanage, S. Yang, X. Liu, Energy and AI 1 (2020).
[45] X. Tang, K. Liu, K. Li, W. D. Widanage, E. Kendrick, F. Gao, Patterns 2 (2021).
[46] Y. Liu, Q. Zhou, G. Cui, Small Methods 5 (2021).
[47] M. Dubarry, D. Beck, J. Power Sources 479 (2020).
[48] Y. Preger, H. M. Barkholtz, A. Fresquez, D. L. Campbell, B. W. Juba, J. Roman-Kustas, S. R. Ferreira, B. Chalamala, J. Electrochem. Soc. 167 (2020).
[49] T. Raj, A. A. Wang, C. W. Monroe, D. A. Howey, Batteries and Supercaps 3 (2020) 1377–1385.
[50] S. Saxena, D. Roman, V. Robu, D. Flynn, M. Pecht, Energies 14 (2021).
[51] X. Hu, Y. Che, X. Lin, S. Onori, IEEE Trans. Transportation Electrification 7 (2021).
[52] D. Roman, S. Saxena, V. Robu, M. Pecht, D. Flynn, Nat. Mach. Intell. 3 (2021) 447–456.
[53] W. Wu, R. Ma, J. Liu, M. Liu, W. Wang, Q. Wang, Int. J. Heat and Mass Transfer 170 (2021).
[54] Z. Jiang, J. Li, Y. Yang, L. Mu, C. Wei, X. Yu, P. Pianetta, K. Zhao, P. Cloetens, F. Lin, Y. Liu, Nat. Commun. (2020).
[55] G. Qian, J. Zhang, S. Chu, J. Li, K. Zhang, Q. Yuan, Z. Ma, P. Pianetta, L. Li, K. Jung, Y. Liu, ACS Energy Lett. 6 (2021) 687–693.
[56] A. Bhowmik, I. E. Castelli, J. M. Garcia-Lastra, P. B. Jorgensen, O. Winther, T. Vegge, Energy Storage Mater. 21 (2019) 446–456.
[57] B. Chen, M. R. Kunz, T. R. Tanim, E. J. Dufek, Cell Reports Phys. Sci. 2 (2021).
[58] M. Dubarry, B. Y. Liaw, J. Power Sources 194 (2009) 541–549.
[59] M. Dubarry, C. Truchot, B. Y. Liaw, J. Power Sources 219 (2012) 204–216.
[60] M. Dubarry, C. Truchot, B. Y. Liaw, J. Power Sources 258 (2014) 408–419.
[61] M. Dubarry, C. Truchot, B. Y. Liaw, K. Gering, S. Sazhin, D. Jamison, C. Michelbacker, J. Power Sources 196 (2012) 10336–10343.
[62] Z. Ma, Z. Wang, R. Xiong, J. Jiang, J. Clean. Prod. 193 (2018) 379–390.
[63] A. Fly, R. Chen, J. Energy Storage 29 (2020).
[64] B. Pan, D. Dong, J. Wang, J. Nie, S. Liu, Y. Cao, Y. Jiang, Electrochim. Acta 362 (2020).
[65] S. Lee, Y. Kim, 2020 American Control Conference Denver, CO, USA (2020).
[66] K. S. Mayilvahanan, K. J. Takeuchi, E. S. Takeuchi, A. C. Marschilok, A. C. West, Batteries and Supercaps 4 (2021).
[67] J. Tian, R. Xiong, W. Shen, F. Sun, Energy Storage Mater. 37 (2021) 283–295.
[68] K. S. Mayilvahanan, J. Kuang, A. H. MacCarthy, L. Wang, K. J. Takeuchi, A. M. Marschilok, E. S. Takeuchi, A. C. West, J. Electrochem. Soc. 168 (2021).
[69] J. Li, D. Wang, L. Deng, Z. Cui, C. Lyu, L. Wang, M. Pecht, J. Energy Storage 31 (2020).
[70] J. Zhu, M. S. D. Darma, M. Knapp, D. R. Sorensen, M. Heere, Q. Fang, X. Wang, H. Dai, L. Meracre, A. Senyshyn, X. Wei, H. Ehrenberg, J. Power Sources 448 (2020).
[71] C. Pastor-Fernandez, K. Uddin, G. H. Chouchelamane, W. D. Widanage, J. Marco, J. Power Sources 360 (2017) 301–318.
[72] H. Sun, B. Jiang, H. You, B. Yang, X. Wang, X. Wei, H. Dai, Energies 14 (2021).
[73] A. G. Li, K. Mayilvahanan, A. C. West, M. Preindl, J. Power Sources 500 (2021).
[74] R. Carter, T. A. Kingston, R. W. Atkinson, M. Parmananda, M. Dubarry, C. Fear, P. P. Mukherjee, C. T. Love, Cell Reports Phys. Sci. 2 (2021).
[75] B. Wu, V. Yufit, Y. Merla, R. F. Martinez-Botas, N. P. Brandon, G. J. Offer, J. Power Sources 273 (2015) 495–501.
[76] K. Liu, T. R. Ashwin, X. Hu, M. Lucu, W. D. Widanage, Renew. Sustain. Energy Rev. 131 (2020).
[77] P. Gasper, K. Gering, E. Dufek, K. Smith, J. Electrochem. Soc. 168 (2021).
[78] W. Li, N. Sengupta, P. Dechent, D. Howey, A. Annaswamy, D. U. Sauer, J. Power Sources 506 (2021).
[79] A. Nuhic, T. Terzimehic, T. Soczka-Guth, M. Buchholz, K. Dietmayer, J. Power Sources 239 (2013) 680–688.
[80] A. Eddahech, O. Briat, N. Bertrand, J. Deletage, J. Vinassa, Electrical Power and Energy Systems 42 (2012) 487–494.
[81] K. Pugalenthi, H. Park, S. Hussain, N. Raghavan, IEEE Access 9 (2021).
[82] Z. Tong, J. Miao, S. Tong, Y. Liu, J. Clean. Prod. 317 (2021).
[83] R. R. Richardson, M. A. Osborne, D. A. Howey, J. Power Storage 357 (2017) 209–219.
[84] R. R. Richardson, M. A. Osborne, D. A. Howey, J. Energy Storage 23 (2019) 320–328.
[85] Y. Choi, S. Ryu, K. Park, H. Kim, IEEE Access 7 (2019) 75143–75152.
[86] M. S. Hosen, R. Youssef, T. Kalogiannis, J. V. Mierlo, M. Berecibar, J. Energy Storage 40 (2021).

[87] Z. Ma, R. Yang, Z. Wang, Appl. Energy 237 (2019) 836–847.
[88] H. Dai, G. Zhao, M. Lin, J. Wu, G. Zhang, IEEE Trans. Ind. Electron. 66 (2019) 7706–7716.
[89] D. Lin, Y. Zhang, X. Zhao, Y. Tang, Z. Dai, Z. Li, X. Wang, G. Geng, J. Energy Engineering 147 (2021).
[90] S. Son, S. Jeong, E. Kwak, J. Kim, K. Oh, Energy 238 (2022).
[91] K. A. Severson, P. M. Attia, N. Jin, N. Perkins, B. Jiang, Z. Yang, M. H. Chen, M. Aykol, P. K. Herring, D. Fraggedakis, M. Z. Bazant, S. J. Harris, W. C. Chueh, R. D. Braatz, Nat. Energy 4 (2019) 383–391.
[92] Z. Fei, F. Yang, K. Tsui, L. Li, Z. Zhang, Energy 225 (2021).
[93] C. Strange, G. dos Reis, Energy and AI 5 (2021).
[94] D. Yang, X. Zhang, R. Pan, Y. Wang, Z. Chen, J. Power Sources 384 (2018) 387–395.
[95] P. Tagade, K. S. Hariharan, S. Ramachandran, A. Khandelwal, A. Naha, S. M. Kolake, S. H. Han, J. Power Sources 445 (2020).
[96] X. Feng, C. Weng, X. He, X. Han, L. Lu, D. Ren, M. Ouyang, IEEE Trans. Vehicular Technology 68 (2019) 8583–8592.
[97] R. R. Richardson, C. R. Birkl, M. A. Osborne, D. A. Howey, IEEE Trans. Ind. Inf. 15 (2019) 127–138.
[98] S. Khaleghi, D. Karimi, S. H. Beheshti, M. S. Hosen, H. Behi, M. Berecibar, J. V. Mierlo, Appl. Energy 282 (2021).
[99] Y. Zhang, Q. Tang, Y. Zhang, J. Wang, U. Stimming, A. A. Lee, Nat. Commun. (2020).
[100] D. Yang, Y. Wang, R. Pan, R. Chen, Z. Chen, Energy Procedia 105 (2017) 2059–2064.
[101] A. G. Li, State Estimation in Lithium-ion Batteries Using Pulse Perturbation and Feedforward Neural Networks, Master's thesis, Columbia University, New York, N.Y., U.S.A., 2020.
[102] Y. A. Fahmy, W. Wang, A. C. West, M. Preindl, J. Energy Storage 41 (2021).
[103] A. Weng, P. Mohtat, P. M. Attia, V. Sulzer, S. Lee, G. Less, A. Stefanopoulou, Joule 5 (2021) 1–22.
[104] W. Weppner, R. A. Higgins, J. Electrochem. Soc. 124 (1977).
[105] C. J. Wen, B. A. Boukamp, R. A. Higgins, J. Electrochem. Soc. 126 (1979).
[106] Y. Zhu, C. Wang, J. Phys. Chem. C 114 (2010) 2830–2841.