

The application of data-driven methods and physics-based learning for improving battery safety

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Abstract

Enabling accurate prediction of battery failure will lead to safer battery systems, as well as accelerating the cell design and manufacturing processes, leading to increased consistency and reliability. Data-driven prediction methods have shown promise for accurately predicting cell behaviors with low computational cost, but they are expensive to train. Furthermore, since the risk of battery failure is already very low, gathering enough relevant data to facilitate data-driven predictions is extremely challenging. Here, a perspective for designing experiments to facilitate a relatively low number of tests, handling the data, applying data-driven methods, and improving our understanding of behavior-dictating physics is outlined. This perspective starts with effective strategies for experimentally replicating rare failure scenarios and thus dramatically reducing the number of experiments, and proceeds to describe means to acquire robust and high-quality datasets, apply data-driven prediction techniques, and to extract physical insights into the events that lead to failure by incorporating physics into data-driven approaches.

Context and Scale

Although hazardous failure of lithium-ion batteries is rare, the fallout can be severe. The safety and reliability of lithium-ion batteries are more important now than ever due to their widespread adoption, yet our ability to predict failure through on-line and off-line diagnostics is still very limited. Lithium-ion batteries are highly complex, nonlinear systems. To make matters worse, two cells of identical geometry, chemistry, and history may respond differently when exposed to identical mechanical, thermal or electrical stimuli. This limits the value of classical deterministic modelling techniques. Applying a probabilistic approach allows for quantification of uncertainty to support decisions in design and control.

Machine learning algorithms are well suited for predicting nonlinear systems like lithium-ion cells, but training and validation of algorithms is challenging for safety applications because large amounts of failure data are needed. Even if the algorithms predict accurately, machine learning is typically agnostic to underlying physics and thus presents limited value in informing researchers and engineers on design opportunities to improve the cells' performance. There is much interest within the battery research community in tackling these challenges, and this perspective aims to offer suggestions on promising avenues of investigation to achieve accurate predictions of the risk of cell failure while gaining some physical insights into the predicted behaviors.

1. Introduction

In engineering, the term ‘safety’ is well described by “the reduction or minimization of risk and uncertainty of harmful events”¹. The important implication behind it is that safety is a qualitative goal, while ‘risk’ is what we can typically quantify and control to achieve that goal². Users of systems that depend on lithium (Li)-ion batteries take on some level of risk that one or more of the Li-ion cells might hazardously fail during the system’s operational lifetime. The risk of hazardous failure of batteries from reputable manufactures is very low but needs to be understood and quantified for regulators and insurers. Depending on the behavior of cells and the operating conditions to which they were exposed during their lifetime, the risk of a cell violently failing varies and changes. Accurately predicting this risk is extremely challenging.

The behavior of Li-ion batteries throughout their lifetime is nonlinear, with a plethora of dynamic electrochemical and mechanical phenomena occurring within the cell at any point in time, whether operating or not. Examples of degradation phenomena include electrolytes reacting with the active materials³, electrode particles degrading through changes in stoichiometry and architecture⁴, and the mechanical properties of separators degrading during cycling⁵, to name a few. Degradation mechanisms are highly dependent on the operating conditions and hence the operational history of the cell is critical to predicting its state and risks. Just as the past and present performance of batteries are used to predict the remaining useful life (RUL) of a battery^{6, 7}, similar historic data may also be used to estimate the risk of a cell as it continues to age. Predicting the likelihood of an internal short circuit however cannot be done with a deterministic model due to the occurrence being somewhat stochastic; behavioral divergence of cells of the same type are known to occur^{8, 9}, which might be linked to subtle variations in synthesis conditions and unintentional defects or contamination occurring during manufacturing. On-line monitoring of a cell’s behavior during its operational lifetime is required to identify when and by how much its behavior has diverged from the expectations^{10, 11} and whether it poses an increased risk of failure (e.g. thermal runaway¹²). On-line data-driven methods have been applied successfully to Li-ion batteries for predicting their RUL¹³⁻¹⁵. Gathering data for predicting typical expected RUL under specific operating conditions is easily achieved and some datasets have even been made public for others to test their algorithms^{14, 16}. However, due to the rarity of catastrophic failure, gathering data which includes outlier cells that eventually undergo thermal runaway may require cycling millions of monitored cells (ideally simultaneously to minimize confounding factors). This may not be as much of a problem for large electric vehicle manufactures who are monitoring billions of deployed cells but it is a major challenge for universities and research institutes.

For gathering enough relevant data, there are opportunities for experimental optimization^{13, 17}, as well as methods for intentionally inducing the rare scenarios that lead to otherwise unpredictable thermal runaway of deployed cells. There may also be opportunities to expand diagnostic techniques to specifically home in on signals that stem from known physical failure mechanisms which would provide insight into how specific phenomena govern degradation. Insights into physical failure mechanisms would also help guide physics-based models and help identify engineering designs for safer battery systems. Hence, it is vital for the battery community to continue to improve physics-based models whilst starting to develop data-driven methods.

There are numerous challenges in achieving accurate data-driven predictions of the risk of battery failure, and in this perspective we discuss possibilities and challenges for designing experiments, handling data,

applying data-driven methods, and improving our understanding of the physical phenomena that lead to failure through physics-based modelling.

Experiment optimization and inducing rare failure scenarios

The types of failure of most interest to detect and predict are those that appear to occur spontaneously during normal operating conditions, the onset of which are difficult to detect. These types of catastrophic failures are rare but known causes exist, such as faulty tabs¹⁸, foreign object debris, welding burrs that press through the separator¹⁹ (Figure 1a), defective placement of electrodes and current collectors²⁰, and gradual mechanical weakening of the separator⁵. The common factor among all the failure mechanisms is that thermal runaway initiates from an internal short circuit which typically occurs from breakage of the separator, whether through mechanical puncture, strain, or weakening of the polymer.

Rare failure scenarios could be emulated through experiments designed to intentionally induce an internal short circuit or weaken the separator through mechanical or thermal means, thus increasing the likelihood of thermal runaway for testing purposes. This can drastically reduce the number of cells required to observe and fully characterize the relevant processes. Placed among normal cells, these high-risk cells would facilitate experiments that use a manageable number of cells to determine whether defective cells can be identified, and whether data-driven methods can predict the likelihood of a cell undergoing thermal runaway and when.

Intentionally inducing an internal defect to simulate an internal short circuit scenario has recently been achieved by incorporating a device inside different cell formats that triggers an internal short circuit on demand, such as by a low melting-point wax²¹ (Figure 1b) or shape memory alloy²² that connect the positive and negative electrodes when moderately elevated temperatures are reached. Both techniques could still be improved by having finer control of the resistance of the short circuit that would empower researchers to control the severity of the short and thus whether the short is likely to induce thermal runaway. The separator could also be made more likely to break by applying force on the cell and straining the separator, through cycling at elevated temperatures²³, or by incorporating a conducting object and lightly pressing on the cell such that the object punctures the separator. Experimental optimization could be applied to achieve a favorable probability of thermal runaway through exploration-exploitation algorithms similar to that recently conducted by Attia et al.¹³, but for cycling conditions and compression or temperature, for example.

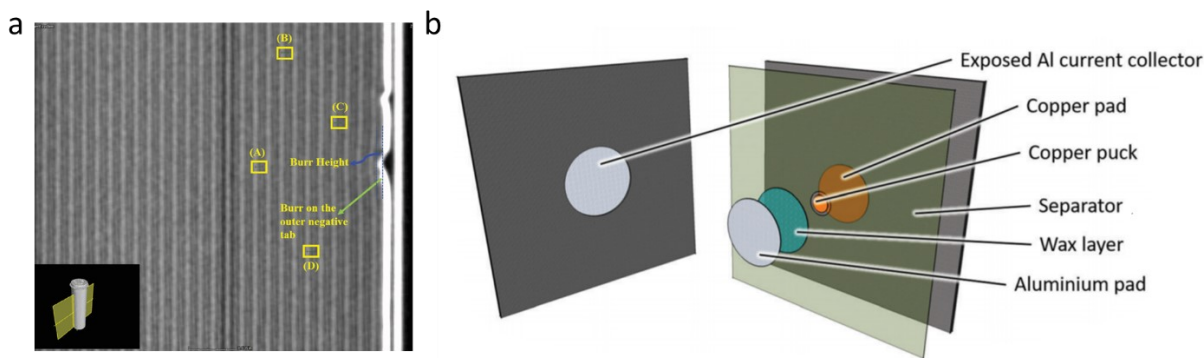


Figure 1. (a) Cross section from an X-ray computed tomography reconstruction of a cylindrical cell showing tab burr protruding into the electrode layers. Reproduced from ¹⁹. (b) Wax-based internal short circuiting device that initiates thermal runaway at moderately elevated temperatures²¹ [*Energy Environ. Sci.*, 2017,10, 1377-1388] - Published by The Royal Society of Chemistry.

However, upon identifying conditions under which thermal runaway is likely, there will be cell-to-cell variation, and for the same cell under the same conditions there will be a spectrum of failure scenarios and risks. For example, separators have been shown to fail in different ways when exposed to similar mechanical loadings and different failure mechanisms can lead to different magnitudes of short circuits²⁴. It is likely that through measuring the response of both normal and stressed cells during cycling, behavioral differences will emerge that may provide some indication of the magnitude of an internal short circuit and the likelihood that the cell will undergo thermal runaway. It is yet to be seen whether the signatures of pre-onset thermal runaway can be effectively detected using available measurement techniques for heat, temperature, acoustic emission, coulombic efficiency, or electrochemical processes. Hence, identifying the expected magnitude of these signals of interest relevant to the onset of hazardous trajectories, and thereafter measuring them, is a crucial next step.

2. Recording data and linking physical phenomena to electrochemical measurements

Until now, data-driven prediction methods have mostly used electrochemical data, which captures degradation from the sum of many physical phenomena but has a limited capability to pinpoint the predominant degradation mechanisms and is not spatially resolved. To assist with gaining physical insight into the system and with interpreting the causes of behavioral divergence, alternative simultaneous operando measurements are needed. Specific accessible operando measurements that can be applied in laboratories and onboard electric vehicles are discussed in the following sections. This section focuses on highlighting means to record signals that provide some physical insight into the system. Figure 2 gives an overview of how signals from cells, including electrochemical, mechanical, acoustic, and thermal, can be used to interpret and perhaps to improve the predictions made by data-driven approaches. Brief descriptions of how these signals are linked to physics are provided below. In field applications such as an electric vehicle, the recorded data can be reported to the cloud, where data-driven analyses could be conducted. The processor on-board can also be used to diagnose problems with the battery in real-time. However, the on-board processor usually has limited resource of data storage and processing, therefore the use of integrated data combined with pretrained models is better suited for on-board use.

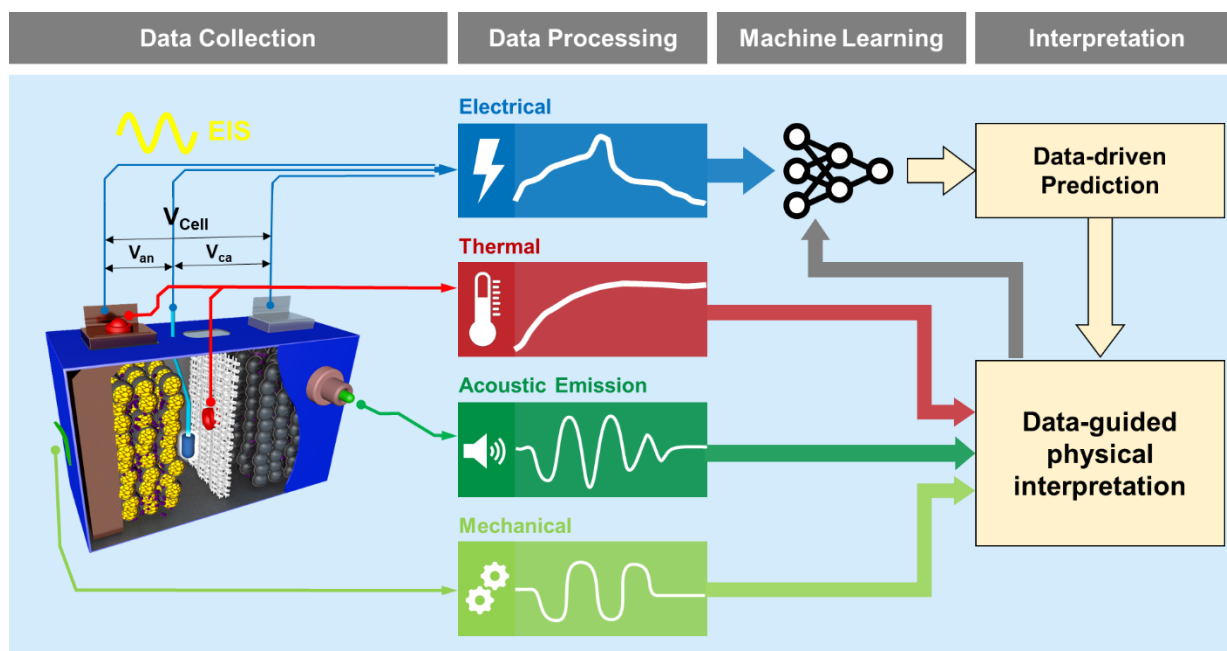


Figure 2. Recording the electrochemical and physical data of a cell for data-driven prediction and physical interpretations.

2.1. Electrochemical behavior

The electrochemical behavior of cells can provide insight into safety issues. For example, abnormal voltage loss can reflect an internal short circuit²⁵ or changes in the voltage plateau during open circuit relaxation can be an indicator of Li plating²⁶. However, voltage under constant current charge/discharge provides insufficient information for inferring the specific degradation mechanisms within the cell. One way to extract more information is using differential analysis by calculating the voltage derivatives²⁷. Sinusoidal current input excites the response of the complex impedance of a cell; this can be conducted for many frequencies through Electrochemical Impedance Spectroscopy (EIS), which can be useful for understanding how a cell's impedance (e.g. limitations in the transportation of Li/Li^{+} ²⁸) evolves. The Distribution of Relaxation Times (DRT)²⁹ approach is a powerful tool for extracting quantitative information on the internal processes occurring in the cell, that are also often easier to interpret graphically. Model-based methods are also helpful to extract fault information from electrical data³⁰. For example, Naha et al.³¹ used a Random Forest (RF) classifier approach in combination with physics motivated equivalent circuit models to predict internal short circuits for mechanically abused cells. Thus, operando electrochemical measurements and simultaneous data processing can facilitate both in-lab and onboard diagnostics for fault recognition and adaptive risk predictions.

2.2. Mechanical behavior

Mechanical behavior that can be indicative of increased risk of a cell failing includes swelling due to gas generation, expansion/contraction due to (de)lithiation, or the cell becoming stiffer due to electrolyte dry-out. Such internal mechanical change could be measured *in operando* by stress or force signals. For example, measurements by force sensors can be used to infer the swelling of Li-ion batteries^{32, 33}. Thin film strain gauges are also useful for measuring cell expansion during cycling³⁴. Acoustic methods have also shown promise in quantifying physical degradation mechanisms and defects. For example, acoustic

time-of-flight analysis has been applied to estimate the state-of-health of batteries³⁵. Acoustic time-of-flight can also reveal information on electrolyte wetting or drying³⁶, and monitoring the acoustic emission from cracking particles may also be a valuable indicator of the cell's degradation rate.

2.3. Thermal behavior

The thermal behavior of a cell is an important metric to monitor when assessing the risk of hazardous failure. The thermal behavior of a cell can reveal anomalies like increasing internal resistances, exothermic reactions, and internal short circuits. Theoretically, abnormal heat generation inside a cell will be reflected in the surface temperature³⁷. However, ideally internal temperatures should also be monitored to detect any thermal deviations accurately. Recently, some minimally invasive techniques have been applied for operando measurement of internal temperatures³⁸. The ohmic resistance and the entropy, which are the two key parameters of heat generation, can be identified online by model-based diagnosing algorithms³⁹. Many of the key parameters for thermal models can be extracted from thermal calorimetry⁴⁰. Spatial surface temperature profiles can also be indicators of underlying defects and can be monitored using thermal imaging⁴¹. There are opportunities for the application of machine learning approaches to extract the battery behavior from many of these physical signals, such as from thermal imaging data⁴².

3. Building a database and improving the accuracy of predictions

As highlighted in the introduction, obtaining representative datasets for quantifying the risk of failure still presents a significant challenge not just within the academic community, but also to manufactures. In the academic literature, it is extremely rare to see studies that investigate more than one hundred of the same cell type, due to resource limitations. Furthermore, cells may not be tested concurrently or for long enough periods to reproduce operational lifetimes, which complicates interpretation. It is notable that a recent article by Severson et al.¹⁴ drew significant attention in the community for exploring a data-driven approach to predicting cycle life. 124 cells were tested, which is large by academic standards, and already an outlier cell was observed exhibiting degradation ten times faster than was typical of the batch. However, although a rapidly degrading cell is undesirable, there's no guarantee that this cell would evolve into a safety risk, but it is generally considered to be a risk factor⁴³. It is also worth noting that although Severson's model (physically motivated feature engineering with linear regression) performed reasonably well for the group, it over-predicted the short-life outlier's cycle life by around 100%.

A study by Chemali et al.⁴⁴ employed a deep-learning approach to try and predict state of charge without the need for feature engineering. Although the power of deep-learning lies in its ability to learn to identify highly complex features from training data, a direct consequence of this is its tendency to over-fit small datasets, which means that they essentially remember a specific dataset rather than extracting the relations that underpin it. Furthermore, it's important to highlight that while ML models can be trained to map complex relationships, they are fundamentally interpolative and cannot be expected to predict behaviors outside of the training data envelope; alternatively a well parameterized physics based model should be able to extrapolate if the correct physics has been selected. Extracting statistically significant results for predicting gradual cell degradation processes is hard enough, but building predictive models of a stochastic binary process (such as internal shorting under normal operating conditions) is far harder. This is why methods have been developed to exacerbate certain failure modes by deliberately operating the cell under stressful conditions, directly damaging the cell, or modifying the cell itself to contain a

known fault⁴⁵. However, it can be difficult to confidently relate these results back to predictions about normal cells under standard operating conditions.

At the commercial scale, rather than trying to simply run very large cell cycling studies in-house, a huge opportunity resides in extracting data from cells during operation. This is of particular relevance to electric vehicle applications as these high-value, safety-critical products contain sufficient onboard instrumentation, telemetry, and computing power to share detailed pack diagnostics and prognostics. Further data-centric approaches by manufactures, such as cell traceability, could help relate lifetime data of all cells produced to every measurement taken during its manufacturing process. While reputable manufacturers generally achieve consistent and high-quality cells, there is always non-zero variance in any manufacturing process. Moving from batch-level traceability to measurement collection for individual cells will further increase the potential for detailed analysis and data-driven modeling. Recycling presents an additional data collection opportunity, where predictions of degradation at end-of-life can be verified.

Figure 3 illustrates a concept for the flow of traceable cells, data and models, between manufacturers, researchers and end-users. By combining the detailed analysis on a small number of cells provided to academia, with (appropriately anonymized) data from many deployed packs, it will be possible to provide not only accurate predictive models, but also insights into the mechanisms causing the behaviors. This will then be fed back to manufactures allowing them to optimize their designs, simulations and manufacturing processes at both cell and system level.

One of the challenges here is that each vehicle will be applying a unique usage pattern to its pack, which makes system-to-system and cell-to-cell comparison more difficult than the equivalent in-house testing under nominally identical conditions. Methods such as rain-counting⁴⁶ exist to try and decompose irregular cycling data into a linear combinations of regular cycling patterns, but these approaches necessarily miss some of the codependences and nonlinearities.

It may be that predictive models of cell failure are too difficult to validate with high confidence, which is why some studies are focusing on data-driven methods for early detection of failure instead, as this could enable mitigation strategies to be deployed by the battery management system (BMS).

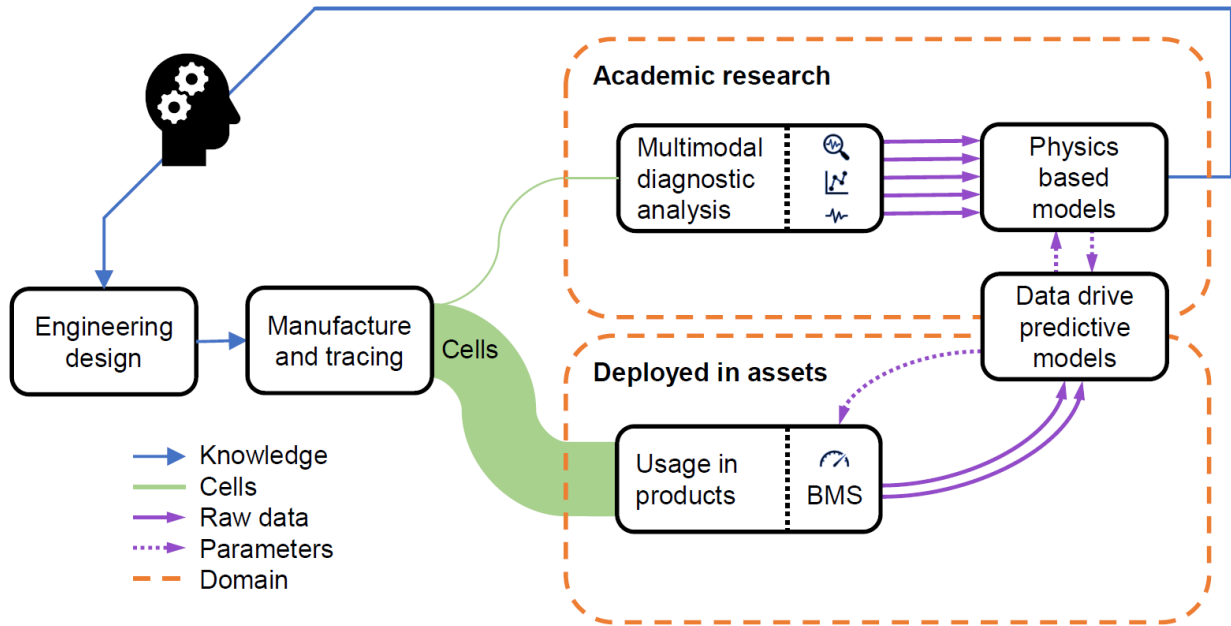


Figure 3. Illustration showing a concept flow of traceable cells, data, and information that leverages field and research data to improve the accuracy of data driven predictions, and the safety and performance of batteries through informed engineering design of cells and battery management systems.

4. Incorporating physics into data-driven predictions and achieving engineering guidance

Data-driven modeling has shown a number of key advantages over its physics-based counterpart⁴⁷⁻⁴⁹, such as substantially reducing the expertise required to use the models. However, purely-data-driven models do not provide much physical insight into the system, which can be somewhat frustrating and unsettling to engineers looking to design safer battery systems. The interpolative nature of many data-driven methods will not only result in poor predictions during extrapolation but will also not flag the uncertainty of these prediction without specifically applying a probabilistic approach. It is important to explore how data-driven methods can be applied to prediction of battery risks while also achieving physical insights for design engineering. The key lies in incorporating physics into the data-driven methods. Here, we identify three key elements from the open literature, as illustrated in Figure 4.

4.1. Physics-based datasets

To promote data collection for engineering guidance, physics-based datasets should be the foundation of any data-driven model related to battery safety. In the previous sections of this Perspective, we elaborated how such a physics-based battery safety dataset can be built through experiments and used to train a machine learning algorithm. It is also worth emphasizing that the predictions of data-driven models should be linked to physical explanations. In most cases, a “testing” procedure is performed. But oftentimes, the testing and the training database are randomly chosen from the same data envelope for convenience, and the testing procedure is consequently interpolative^{50,51}. A stricter criterion for validation is whether the model is able to provide more insights into the physics, referred to as extrapolative. A typical example of such extrapolative validations in battery research is the prediction of remaining useful

lifetime (RUL) of battery cells using the measured data during the service life^{11, 52}. Various data-driven approaches based on machine learning, statistical analysis, and signal processing have been systematically summarized in several review articles^{4, 6, 53}. Another example related to battery safety is identifying the most critical condition of battery cells under which a mechanical failure or short circuit is most likely to happen¹⁷. It is usually difficult to perform experiments to find such conditions, and the challenge is how to extrapolate the directly measurable dataset to the unknown area. Identifying the most critical condition is of great real-world value for the automotive industry to reduce the size of the test matrix, standardize the test condition, and guide the passive safety design of electric vehicles.

In many cases, a large experimental dataset is usually unaffordable or inaccessible, for example, the investigation of the nanoscale and atomic-level structure-property relation. Databases generated by high-throughput first-principal computations are thus becoming increasingly important to be used in training train neural networks. In a recent study, Chen et al.⁵⁴ predicted the phonon density of states of crystalline solids, using a density functional perturbation theory calculated phonon database⁵⁵, to train a Euclidean neural network. The algorithm has a good extrapolative capability to predict the properties of unseen crystal structures for the discovery of materials with high specific heat capacity. Similar successes have been achieved in searching for next generation energy materials, as summarized in a recent review article⁵⁶.

4.2. Physics-informed training constraints

Li-ion batteries are complex systems involving multiple physics, scales, and phases. Each of the aspects can be described by a set of governing equations, usually partial differential equations (PDEs), such as the heat and mass transfer equations as well as balances of mechanical force and momentum. In most physics-based models or first-principle-based models, these equations with proper initial and boundary conditions are already identified by existing knowledge, and the task of the model is to solve them. Conventional purely-data-driven methods are oftentimes supposed to bypass solving the governing equations and directly provide a solution. Consequently, the prediction may violate some basic laws of physics.

Two promising approaches of incorporating the physical governing equations into the data-driven framework are (1) reformulating the models with mathematical techniques such as Galerkin and Spectral methods to reduce the computational cost, and (2) using data-driven approach to solve highly nonlinear differential equations. Here in this perspective, we offer a brief overview of the second approach, which has accumulated a significant number of early successes over the past two years. The essence of this approach is incorporating the nonlinear equations, as well as their boundary and initial conditions, into the data-driven model as training constraints (referred to as “physics-informed training constraints” hereinafter). Several examples are the Physics Informed Neural Network (PINN) algorithm developed by Raissi et al.^{57, 58}, the machine learning-integrated first-principle-based modeling framework established by Zhang et al.^{59, 60}, the deep learning library of solving commonly-seen differential equations by Lu et al.⁶¹, and the PDE-constrained optimization method by Zhao et al.⁶². A practical application of these algorithms in battery safety modeling is providing an accurate approximate solution of the multiphysics PDEs to bypass some issues that cannot be effectively addressed by conventional computational methods. One of the issues is the reliance on a high-quality mesh for finite element methods. In many battery failure simulations, large local deformation and crack propagation happen. Electrochemical PDEs are usually defined on interfaces involving mass transport and growth. Creating a robust mesh for these problems is

difficult and expensive. Many deep learning approaches⁶³⁻⁶⁵ for solving PDEs could be mesh-free and, therefore, have a great potential to be used in the multiphysics modeling of batteries.

One more clear advantage of such physics-informed data-driven approaches is that the cost of obtaining a sufficiently large database for training can be tremendously decreased. For example, in both algorithms developed by Raissi et al.^{57, 58} and by Lu et al.⁶¹, the boundary conditions and initial conditions of the PDEs are used as the loss function to train the network parameter (as illustrated in Figure 4b), which are relatively cheap to obtain compared with data points at arbitrary special coordinates with a complex time history. These algorithms turned out to be highly successful to solve a wide class of nonlinear PDEs, and their application in energy storage systems is quite promising. In particular, the ability to use only a small set of images⁶² to learn advanced continuum models of Li-ion batteries based on nonequilibrium thermodynamics⁶⁶ could be revolutionary, if applied to *operando* image data for ion intercalation in primary particles⁶⁷ or porous electrodes⁶⁸. Armed with accurate models of intercalation kinetics, the same approach of image inversion can then be used to learn the physics of degradation processes, such as lithium plating and solid electrolyte interphase (SEI) growth^{69, 70} on carbon anodes, for cell-level simulations using multiphase porous electrode theory (MPET)⁷¹ to enable data-driven, physics-based prediction of battery safety and lifetime.

4.3. Physics-guided algorithm structures

The third element that could be incorporated with physics is the structure in the machine learning algorithm itself. Here, we take the artificial neural network (ANN) approach as an example. It is quite common that a neural network is specially designed to handle a specific problem or physics field. For example, the well-known convolutional neural network was originally developed for image processing and the recurrent neural network is suitable for processing sequential signals. To develop data-driven models of a complex system with multiple physical fields, we shall also consider designing and optimizing the structure of the algorithm under the guidance of physics. Han et al.⁷² proposed a network architecture for solving semi-linear parabolic PDEs, which consist of several subnetworks for different time intervals and the connections between them are established based on the equations to be solved (as illustrated in Figure 4c). A similar approach of algorithm structural design can be applied to the multi-physics and multi-scale problems. A neural network with several subnetworks for different fields and the connections between one another to reflect the real physics behind them can be effective for potential battery safety modeling.

It is worth noting that the three key elements that we identified here should not be understood as a strict classification. There is no absolute boundary among various data-driven approaches. With the rapid development of machine learning technologies, we are witnessing an increasing number of advanced models that have all the above three merits. For example, in a recent study, Qian et al.⁷³ developed a successful framework named as *Lift & Learn*, which turns out to be effective for use as a physics-informed approach to learn low-dimensional models of large-scale dynamical systems. The potential applications of such types of new algorithms in battery failure diagnostics is promising.

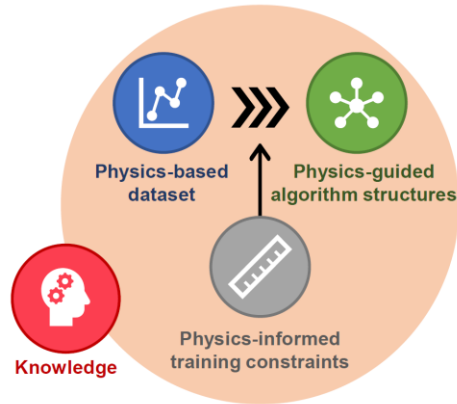


Figure 4. Three key elements of the data-driven framework into which physical can be incorporated. a) Creating a physics-based dataset, b) applying physics-informed training constraints, and c) designing physics-guided algorithm structures.

5.4 Practical implementation for commercial applications

Beyond the three approaches described above, there is also an opportunity for using both measured real-world data and physics-based simulations to synergistically train ML models. Inspiration for this can be taken from the robotics and automation literature. The task of learning robot control in complex systems is challenging due to the “sim-to-real” mismatch. For example, the optimal control policy to achieve upright walking in simulation might fail in the real world due to mismodeled actuators or dynamics. Much as in the battery case, collecting large datasets exclusively from real-world experiments would be prohibitively expensive. Hwasser et al.⁷⁴ has shown that building a conditional Variational Autoencoder (VAE) using real-world experimental measurements and a simulator as “informed regularizer” results in a data-efficient model that identifies the parameter posterior for the simulator to match reality. In addition, the learning process also produces a generative model that acts as a stochastic simulator, outperforming traditional methods using conditional VAEs using only 1-10% of the data. Autoencoders work by compressing a large feature space into a compact, low-dimensional representation (latent space). The values of this latent space tensor are typically not interpretable, but can be forced to be semantically meaningful (e.g. a subset of simulator parameters). The approach successfully implemented in the robotics application starts with real-world experimental data collection with unknown physical parameters (e.g. centre of gravity, coefficient of friction). The same scenario is simulated using a deterministic simulator with a distribution of physical input parameters. The simulator trajectories (initial and final states) are passed alongside the corresponding simulator parameters to train the autoencoder’s decoder whose weights are subsequently frozen before using the experimental data to train the encoder part (**Error! Reference source not found.**). The freezing of weights forces the latent space to represent the semantically meaningful simulation parameters. More data can be collected as deemed necessary, either through real-world experiments, or through simulation (with updated parameters from the autoencoder).

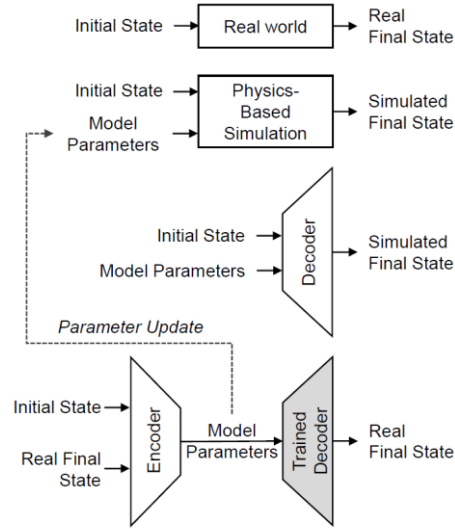


Figure 5. Schematic overview of the application of VAEs to enhanced data-driven model building.

For a Li-ion cell application, the real-world experimental data could be from a laboratory cyclor, as well as the numerous aforementioned measurable signals that relate to the state of the physical system within the cell. The simulation data can be obtained from a high-fidelity physics-based model including degradation mechanisms. The state must be physically measurable e.g. discharge capacity, DC-IR, but not necessarily easily interpretable by humans (e.g. complex load profile response). The parameters will be chosen as physical simulation parameters of interest. If the simulation is of sufficient quality, the approach will converge and give multiple benefits. First, it will be possible to replace simulations by the trained neural network which will offer substantial speed increase. Second, it will be possible to discover the physical parameters that govern the observation of a real-world experiment and thereby provide physical interpretability. In a further step, perhaps the physical parameters can be predicted from design and/or manufacture. If so, it would be possible to chain predictions to run experiment-informed simulations on cells that do not exist.

The approach described above has been hugely impactful on the robotics community, but so far unreported in the battery literature. This presents a major opportunity to not only make use of the deep expertise employed in developing physics-based models, but also to harness the data in an interpretable and robust way. For example, the probability of battery failure following mechanical abuse could be predicted by combining physics-based mechanical models with real-world measurements from the cells, in a stochastic framework. Additionally, this method could be applied to predicting the likelihood of battery failure following an aggressive cycling regime, where electrochemical models and physical signals from the cells are used to simulate the risk of the cell undergoing failure.

Summary and outlook

There are numerous challenges that face the application of data-driven methods for improving battery safety. Notably, the collection of high-quality and robust experimental data currently limits validation and

guidance of algorithms, and even with data, extracting physical explanations from data-driven predictions presents a further challenge. For large battery manufacturers, datacentric initiatives with cell traceability from synthesis to end of life, will become invaluable. This data will facilitate development of robust data-driven predictions of battery behaviors and failure risks, as well as identifying their influencing factors during manufacture. For research laboratories that do not have access to such large databanks, this challenge is far greater. Informed design of experiments to reproduce the rare failure scenarios of interest are needed, such as intentionally inducing an internal short circuit.

Extracting physical explanations from data-driven predictions to help guide design of safer cells and operating conditions requires parallel development of physics-based models. Experimentally, this link to physical phenomena may come from effective *operando* data collection practices that record signals from known physical degradation mechanisms, such as electrochemical, thermal, mechanical, and acoustic monitoring. Some methods like mechanical and acoustic monitoring may not be sufficiently mature, cost effective, or practical to implement on electric vehicles, but if the data is revealed to be sufficiently valuable, miniaturization and cost reduction of the necessary equipment to achieve on-board monitoring is possible. Syncing the signals from physical degradation mechanisms with the electrical data used for predictions, is expected to help link the predicted behaviors to physical phenomena. Furthermore, a researcher could link data-driven predictions back to physics via other *operando* measurements and by physics-based modelling, or they could let the physics guide the machine learning algorithms, increasing the confidence in the results while reducing the quantity of training data required. Finally, further promotion of data-driven methods for improving battery safety would come from all groups making their data open-source for the collective benefit of the global research community through access to robust, plentiful, and high-quality datasets.

Author contributions

All authors contributed to writing the manuscript.

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