CellPress

Joule

Perspective

The Application of Data-Driven Methods and Physics-Based Learning for Improving Battery Safety

Donal P. Finegan,[1,*] Juner Zhu,[2,3] Xuning Feng,[4] Matt Keyser,[1] Marcus Ulmefors,[5] Wei Li,[2] Martin Z. Bazant,[3,7] and Samuel J. Cooper[6]

SUMMARY

Enabling accurate prediction of battery failure will lead to safer battery systems, as well as accelerating cell design and manufacturing processes for 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, given that 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 reducing the number of experiments, and proceeds to describe means to acquire 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.

INTRODUCTION

In engineering, the term "safety" is well described as "the reduction or minimization of risk and uncertainty of harmful events." [1] The important implication behind this definition is that safety is a qualitative goal, whereas "risk" is what we can typically quantify and control to achieve that goal.[2] Users of systems who 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 are 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 active materials,[3] electrode particles degrading through changes in stoichiometry and architecture,[4] and the mechanical properties of separators degrading during cycling,[5] 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

Context & Scale

Although the 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 because of their widespread adoption, yet our ability to predict failure through online and offline 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 might respond differently when exposed to identical mechanical, thermal, or electrical stimuli. This limits the value of classical deterministic modeling 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 are 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 performances of batteries are used to predict the remaining useful life (RUL) of a battery,[6,7] similar historic data might 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 because of 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. Online 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 runaway).[12] Online data-driven methods have been applied successfully to Li-ion batteries for predicting their RUL.[13–15] Gathering data for predicting a typical expected RUL under specific operating conditions are easily achieved and some datasets have even been made public for others to test their algorithms such as thodesse.[14,16] Recently, NREL and NASA also released an open-source dataset called the Battery Failure Databank.[17] However, due to the rarity of catastrophic failure, gathering data that include outlier cells that eventually undergo thermal runaway might require cycling millions of monitored cells (ideally simultaneously to minimize confounding factors). This might not be as much of a problem for large electric vehicle manufactures who are monitoring billions of deployed cells but is a major challenge for universities and research institutes.

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.

For gathering enough relevant data, there are opportunities for experimental optimization,[13,18] as well as methods for intentionally inducing rare scenarios that lead to the otherwise unpredictable thermal runaway of deployed cells. There might 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. Therefore, it is vital for the battery community to continue to improve physics-based models while 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 modeling.

EXPERIMENT OPTIMIZATION AND INDUCING RARE FAILURE SCENARIOS

Failures that occur unexpectedly during normal operation are of most interest to detect and predict, the onset of which is difficult to detect. These types of catastrophic failures are rare but known causes exist, such as faulty tabs,[19] foreign object debris, welding burrs that press through the separator[20] (Figure 1A), defective placement of electrodes and current collectors,[21] and gradual mechanical weakening of the separator.[5] The common factor among all the failure mechanisms is that the thermal runaway initiates from an internal short circuit that typically occurs from breakage of the separator, whether through the 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

[1]National Renewable Energy Laboratory, Golden, CO 80401, USA
[2]Department of Mechanical Engineering, MIT, 77 Massachusetts Ave., Cambridge, MA 02139, USA
[3]Department of Chemical Engineering, MIT, 77 Massachusetts Ave., Cambridge, MA 02139, USA
[4]State Key Laboratory of Automotive Safety and Energy, Tsinghua University, Beijing 100084, China
[5]Northvolt AB, 111 20 Stockholm, Sweden
[6]Dyson School of Design Engineering, Imperial College London, London, UK
[7]Department of Mathematics, MIT, 77 Massachusetts Ave, Cambridge, MA 02139, USA
*Correspondence: donal.finegan@nrel.gov
https://doi.org/10.1016/j.joule.2020.11.018

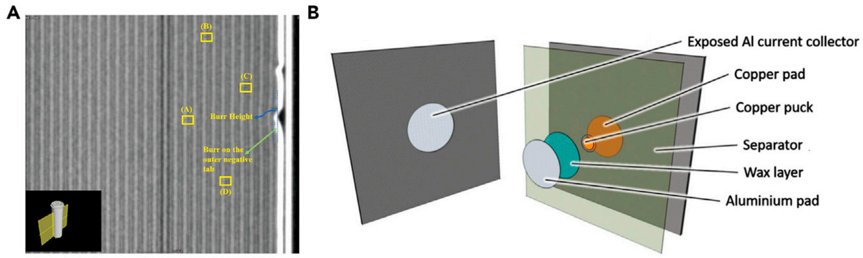

Figure 1. Unintentional and Intentional Causes of Internal Short Circuits

(A) Cross-section from an X-ray computed tomography reconstruction of a cylindrical cell showing tab burr protruding into the electrode layers. Reproduced with permission from IEEE from Yao et al.[20]

(B) Wax-based internal short-circuiting device that initiates thermal runaway at moderately elevated temperatures.[22] Reproduced from [*Energy Environ. Sci.*, 2017,10, 1377–1388] - Published by the Royal Society of Chemistry.

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 wax[22] (Figure 1B) or shape memory alloy[23] 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 temperatures,[24] 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 those recently conducted by Attia et al.,[13] but for cycling conditions and compression or temperature, for example.

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 circuits.[25] It is likely that through measuring the response of both normal and stressed cells during cycling, behavioral differences will emerge that might 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 by using available measurement techniques for heat, temperature, acoustic emission, coulombic efficiency, or electrochemical processes. Hence, identifying the expected magnitude of these signals of interest

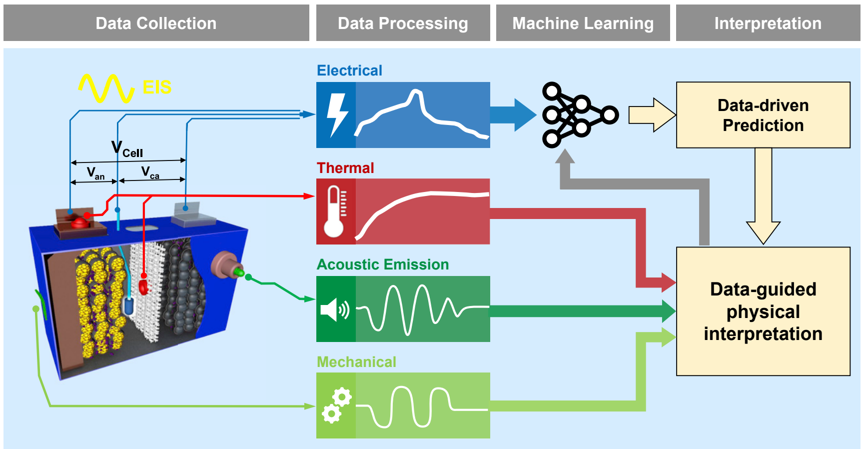

Figure 2. Recording the Electrochemical and Physical Data of a Cell for Data-Driven Prediction and Physical Interpretations

relevant to the onset of hazardous trajectories, and thereafter measuring them, is the crucial next step.

RECORDING DATA AND LINKING PHYSICAL PHENOMENA TO ELECTROCHEMICAL MEASUREMENTS

Until now, data-driven prediction methods have mostly used electrochemical data, which capture degradation from the sum of many physical phenomena but have a limited capability to pinpoint the predominant degradation mechanisms and are not spatially resolved. Alternative simultaneous operando measurements are needed to help gain physical insight into the system and to interpret the causes of behavioral divergence. Specific accessible operando measurements that can be applied in laboratories and on-board 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 can 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 a limited resource of data storage and processing; therefore, the use of integrated data combined with pre-trained models is better suited for on-board use.

Electrochemical Behavior

The electrochemical behavior of cells can provide an insight into safety issues. For example, abnormal voltage loss can reflect an internal short circuit,[26] or changes in the voltage plateau during open-circuit relaxation can be an indicator of Li plating.[27] However, voltage under constant current charge/discharge provides insufficient information to infer the specific degradation mechanisms within the cell. One way to extract more information is by using differential analysis by calculating the voltage derivatives.[28] 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^{+}[29])

evolves. The distribution of relaxation times (DRT)[30] approach is a powerful tool to extract 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 data.[31] For example, Naha et al.[32] 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 on-board diagnostics for fault recognition and adaptive risk predictions.

Mechanical Behavior

Mechanical behavior that can be indicative of an increased risk of a cell failing includes swelling due to gas generation, expansion or 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.[33,34] Thin-film strain gauges are also useful for measuring cell expansion during cycling.[35] 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 batteries.[36] This analysis can also reveal information on electrolyte wetting or drying,[37] and monitoring the acoustic emission from cracking particles may also be a valuable indicator of the cell's degradation rate.

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 such as increasing internal resistances, exothermic reactions, and internal short circuits. Theoretically, abnormal heat generation inside a cell will be reflected in the surface temperature.[38] 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 temperatures.[39] The ohmic resistance and the entropy, which are the two key parameters of heat generation, can be identified online by model-based diagnosing algorithms.[40] Many of the key parameters for thermal models can be extracted from thermal calorimetry.[41] Spatial surface temperature profiles can also be indicators of underlying defects and can be monitored by using thermal imaging.[42] There are opportunities for the application of machine-learning approaches to extract the battery behavior from many of these physical signals, such as those from thermal imaging data.[43]

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 because of resource limitations. Furthermore, cells might 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. A total of 124 cells were tested, which is large according to academic standards, and an outlier cell was observed exhibiting degradation ten times faster than the degradation that 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 is generally considered to be a risk factor.[44] 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.[45] employed a deep-learning approach to try and predict the 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 is important to highlight that, although machine-learning 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 difficult enough but building predicative models of a stochastic binary process (such as internal shorting under normal operating conditions) is far more difficult. 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 fault.[46] However, it can be difficult to confidently relate these results back to predictions about normal cells under standard operating conditions.

On a 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 on-board instrumentation, telemetry, and computing power to share detailed pack diagnostics and prognostics. Further data-centric approaches by manufacturers, such as cell traceability, could help relate lifetime data of all cells produced to every measurement taken during its manufacturing process. Although reputable manufacturers generally achieve consistent and high-quality cells, there is always a 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 levels.

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 comparisons more difficult than the equivalent in-house testing under nominally identical conditions. Methods such as rain counting[47] exist to try and decompose irregular cycling data into linear combinations of regular cycling patterns, but these approaches necessarily miss some of the codependences and nonlinearities.

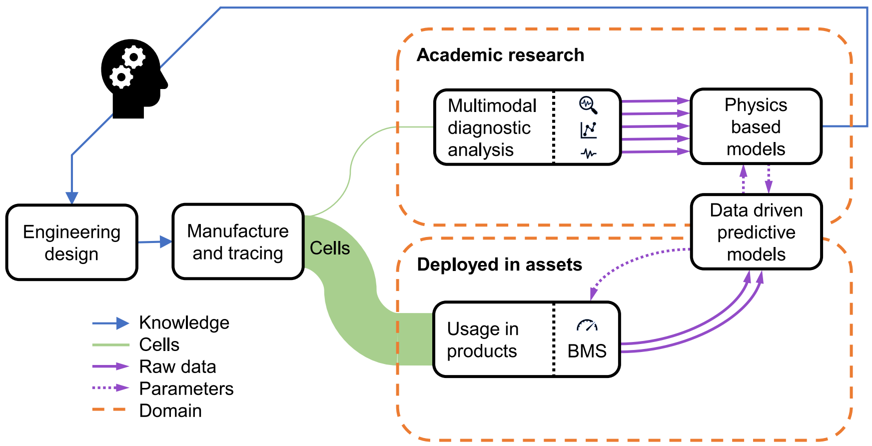

Figure 3. Illustration Showing a Concept Flow of Traceable Cells, Data, and Information that Leverages Field and Research Data to Improve the Accuracy of Data-Driven Predictions, and the Safety and Performance of Batteries through Informed Engineering Design of Cells and Battery Management Systems

It could 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, given that this can enable mitigation strategies to be deployed by the battery management system (BMS).

INCORPORATING PHYSICS INTO DATA-DRIVEN PREDICTIONS AND ACHIEVING ENGINEERING GUIDANCE

Data-driven modeling has shown a number of key advantages over its physics-based counterpart,[48–50] 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 predictions without specifically applying a probabilistic approach. It is important to explore how data-driven methods can be applied to the 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.

Physics-Based Datasets

To promote data collection for engineering guidance, physics-based datasets should be the foundation for 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.[51,52] 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 RUL of battery cells by using measured data during the service life.[11,53] Various data-driven approaches based on machine

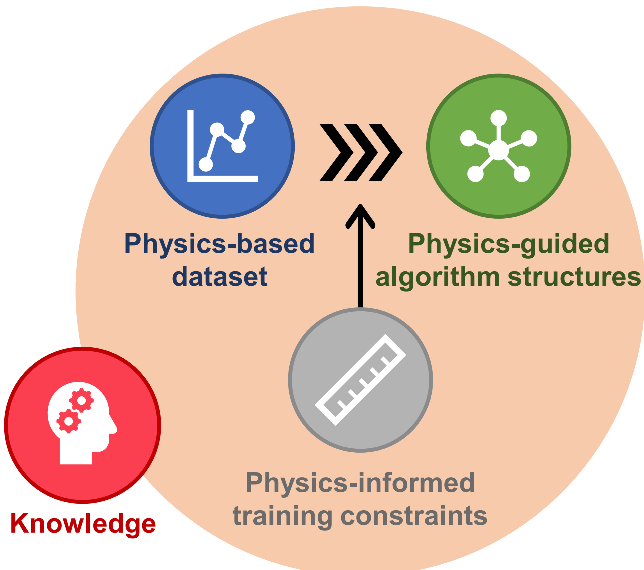

Figure 4. Three Key Elements of the Data-Driven Framework into Which Physics and Knowledge Can be Incorporated

Creating a physics-based dataset, applying physics-informed training constraints, and designing physics-guided algorithm structures.

learning, statistical analysis, and signal processing have been systematically summarized in several review articles.[4,6,54] 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 happen.[18] 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-principle computations are thus becoming increasingly important to be used in training neural networks. In a recent study, Chen et al.[55] predicted the phonon density of states of crystalline solids by using a density functional perturbation theory calculated phonon database[56] 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 a high specific heat capacity. Similar successes have been achieved in searching for next-generation energy materials, as summarized in a recent review article.[57]

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 might 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 a 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 2 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). A few examples to quote are the physics-informed neural network (PINN) algorithm developed by Raissi et al.,[58,59] the machine-learning-integrated first-principle-based modeling framework established by Zhang et al.,[60,61] the deep-learning library of solving commonly seen differential equations by Lu et al.,[62] and the PDE-constrained optimization method by Zhao et al.[63] 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 deformations and crack propagations 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 approaches[64–66] 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.[58,59] and by Lu et al.,[62] the boundary and initial conditions of the PDEs are used as the loss function to train the network parameter (as illustrated in Figure 4), which are relatively cheaper 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 images[63] to learn advanced continuum models of Li-ion batteries based on nonequilibrium thermodynamics[67] could be revolutionary, if applied to *operando* image data for ion intercalation in primary particles[68] or porous electrodes.[69] 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[70,71] on carbon anodes, for cell-level simulations using multiphase porous electrode theory (MPET)[72] to enable data-driven, physics-based prediction of battery safety and lifetime.

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 physical phenomena. 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 phenomena, we shall also consider designing and optimizing the structure of the algorithm under the guidance of physics. Han et al.[73] 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 on the basis of the equations to be solved (as illustrated in Figure 4). A similar approach of algorithm structural design can be applied to the multiphysics and multiscale 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.[74] 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.

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 machine-learning models. Inspiration for this can be taken from the robotics and automation literature. The task of learning robot control in complex systems is challenging because of the "sim-to-real" mismatch. For example, the optimal control policy to achieve upright walking in simulation might fail in the real world because of 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.[75] have shown that building a conditional variational autoencoder (VAE) by using real-world experimental measurements and a simulator as an "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 by 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., the center of gravity, coefficient of friction). The same scenario is simulated by 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 (Figure 5). 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).

For a Li-ion cell application, the real-world experimental data can be obtained from a laboratory cycler, 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.

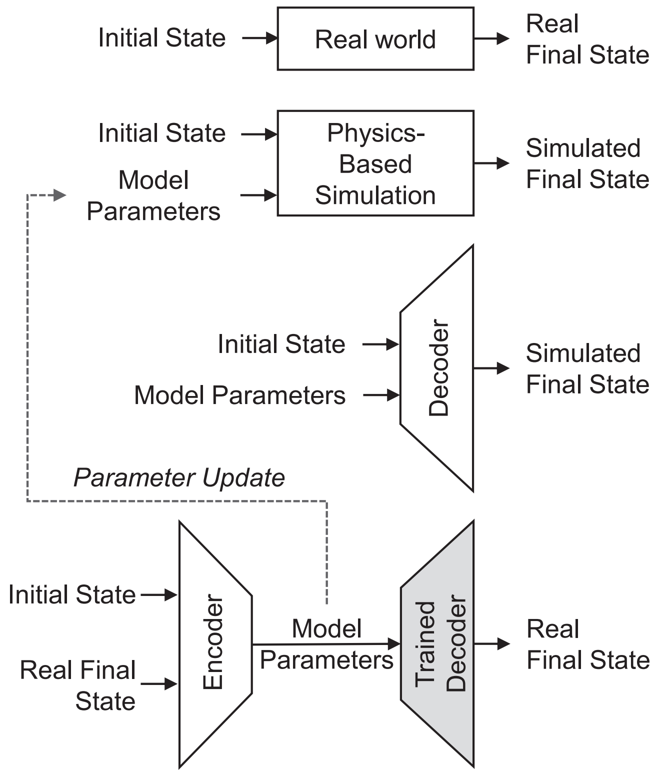

Figure 5. Schematic Overview of the Application of VAEs to Enhance Data-Driven Model Building

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 with the trained neural network, which will offer a 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 after mechanical abuse can be predicted by combining physics-based mechanical models with real-world measurements from the cells, in a stochastic framework. Additionally, this method can be applied to predict the likelihood of battery failure after 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 the validation and guidance of algorithms, and even with data, extracting physical explanations from data-driven predictions presents a further challenge. For large battery manufacturers, data-centric initiatives with cell traceability from synthesis to end of life will become invaluable. These data will facilitate the development of robust data-driven predictions of battery behaviors and failure risks, as well as in 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 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 the design of safer cells and operating conditions requires parallel development of physics-based models. Experimentally, this link to physical phenomena might 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 such as mechanical and acoustic monitoring might not be sufficiently mature, cost-effective, or practical to implement on electric vehicles, but if the data are 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 modeling, 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.

ACKNOWLEDGMENTS

This work was authored by Alliance for Sustainable Energy, LLC, the manager and operator of the National Renewable Energy Laboratory for the U.S. Department of Energy (DOE) under contract number DE-AC36-08GO28308. Funding was provided by the U.S. Department of Energy Vehicle Technology Office. J.Z. thanks the financial support from the MIT Industrial Battery Consortium, the USAID (SHERA) project, and the MIT-Indonesia Seed Fund. X.F. gives thanks for the funding from the National Science Foundation of China under the contract number 51706117, and the support of "Young Elite Scientist Sponsorship Program" from China Association for Science and Technology (2018QNRC001). M.Z.B. acknowledges support from the Toyota Research Institute through the D3BATT center for Data-Driven Design of Rechargeable Batteries. S.J.C. was supported by the EPSRC Faraday Institution Multi-Scale Modeling project (EP/S003053/1, grant number FIRG003).

AUTHOR CONTRIBUTIONS

All authors contributed to writing the manuscript.

REFERENCES

1. Varshney, K. (2016). Engineering safety in machine learning. 2016 Information Theory and Applications Workshop (ITA), pp. 1–5.

2. Moller, N. (2012). The concepts of risk and safety. In Handbook of Risk Theory: Epistemology, Decision Theory, Ethics, and Social Implications of Risk (Springer), pp. 55–85.

3. Henschel, J., Horsthemke, F., Stenzel, Y.P., Evertz, M., Girod, S., Lürenbaum, C., Kösters, K., Wiemers-Meyer, S., Winter, M., and Nowak, S. (2020). Lithium ion battery electrolyte degradation of field-tested electric vehicle battery cells – a comprehensive analytical study. J. Power Sources *447*, 227370.

4. Pender, J.P., Jha, G., Youn, D.H., Ziegler, J.M., Andoni, I., Choi, E.J., Heller, A., Dunn, B.S., Weiss, P.S., Penner, R.M., and Mullins, C.B. (2020). Electrode degradation in lithium-ion batteries. ACS Nano *14*, 1243–1295.

5. Zhang, X., Zhu, J., and Sahraei, E. (2017). Degradation of battery separators under charge–discharge cycles. RSC Adv. 7, 56099–56107.

6. Li, Y., Liu, K., Foley, A.M., Zülke, A., Berecibar, M., Nanini-Maury, E., Van Mierlo, J., and Hoster, H.E. (2019). Data-driven health estimation and lifetime prediction of lithium-ion batteries: a review. Renew. Sustain. Energy Rev. 113, 109254.

7. Hu, X., Xu, L., Lin, X., and Pecht, M. (2020). Battery lifetime prognostics. Joule 4, 310–346.

8. Schuster, S.F., Brand, M.J., Berg, P., Gleissenberger, M., and Jossen, A. (2015). Lithium-ion cell-to-cell variation during battery electric vehicle operation. J. Power Sources 297, 242–251.

9. Harris, S.J., Harris, D.J., and Li, C. (2017). Failure statistics for commercial lithium ion batteries: a study of 24 pouch cells. J. Power Sources 342, 589–597.

10. Saxena, S., Kang, M., Xing, Y., and Pecht, M. (2018). Anomaly detection during lithium-ion battery qualification testing. In 2018 IEEE International Conference on Prognostics and Health Management (ICPHM), pp. 1–6.

11. Cripps, E., and Pecht, M. (2017). A Bayesian nonlinear random effects model for identification of defective batteries from lot samples. J. Power Sources 342, 342–350.

12. Finegan, D.P., and Cooper, S.J. (2019). Battery safety: data-driven prediction of failure. Joule 3, 2599–2601.

13. Attia, P.M., Grover, A., Jin, N., Severson, K.A., Markov, T.M., Liao, Y.H., Chen, M.H., Cheong, B., Perkins, N., Yang, Z., et al. (2020). Closed-loop optimization of fast-charging protocols for batteries with machine learning. Nature 578, 397–402.

14. Severson, K.A., Attia, P.M., Jin, N., Perkins, N., Jiang, B., Yang, Z., Chen, M.H., Aykol, M., Herring, P.K., Fraggedakis, D., et al. (2019). Data-driven prediction of battery cycle life before capacity degradation. Nat. Energy 4, 383–391.

15. Ng, M.-F., Zhao, J., Yan, Q., Conduit, G.J., and Seh, Z.W. (2020). Predicting the state of charge and health of batteries using data-driven machine learning. Nat. Mach. Intell. 2, 161–170.

16. Saha, B., and Goebel, K. (2007). Battery data set. NASA Ames Research Center, Moffett Field CA: NASA Ames Prognostic Data Repository. https://ti.arc.nasa.gov/tech/dash/groups/pcoe/prognostic-data-repository/.

17. Finegan, et al.. (2020). Battery Failure Databank. National Renewable Energy Laboratory. https://www.nrel.gov/transportation/battery-failure.html.

18. Li, W., Zhu, J., Xia, Y., Gorji, M.B., and Wierzbicki, T. (2019). Data-driven safety envelope of lithium-ion batteries for electric vehicles. Joule 3, 2703–2715.

19. Sun, Y., Kong, L., Abbas Khan, H.A., and Pecht, M.G. (2019). Li-ion Battery Reliability – a case study of the Apple iPhone®. IEEE Access 7, 71131–71141.

20. Yao, X., Saxena, S., Su, L., and Pecht, M.G. (2019). The explosive nature of tab burrs in Li-Ion batteries. IEEE Access 7, 45978–45982.

21. Wu, Y., Saxena, S., Xing, Y., Wang, Y., Li, C., Yung, W., and Pecht, M. (2018). Analysis of manufacturing-induced defects and structural deformations in lithium-ion batteries using computed tomography. Energies 11, 925.

22. Finegan, D.P., Darcy, E., Keyser, M., Tjaden, B., Heenan, T.M.M., Jervis, R., Bailey, J.J., Malik, R., Vo, N.T., Magdysyuk, O.V., et al. (2017). Characterising thermal runaway within lithium-ion cells by inducing and monitoring internal short circuits. Energy Environ. Sci. 10, 1377–1388.

23. Zhang, M., Du, J., Liu, L., Stefanopoulou, A., Siegel, J., Lu, L., He, X., Xie, X., and Ouyang, M. (2017). Internal short circuit trigger method for lithium-ion battery based on shape memory alloy. J. Electrochem. Soc. 164, A3038–A3044.

24. Leng, F., Tan, C.M., and Pecht, M. (2015). Effect of temperature on the aging rate of Li ion battery operating above room temperature. Sci. Rep. 5, 12967.

25. Zhang, X., Sahraei, E., and Wang, K. (2016). Li-ion battery separators, mechanical integrity and failure mechanisms leading to soft and hard internal shorts. Sci. Rep. 6, 32578.

26. Feng, X., Pan, Y., He, X., Wang, L., and Ouyang, M. (2018). Detecting the internal short circuit in large-format lithium-ion battery using model-based fault-diagnosis algorithm. J. Energy Storage 18, 26–39.

27. Yang, X.-G., Ge, S., Liu, T., Leng, Y., and Wang, C.-Y. (2018). A look into the voltage plateau signal for detection and quantification of lithium plating in lithium-ion cells. J. Power Sources 395, 251–261.

28. Feng, X., Merla, Y., Weng, C., Ouyang, M., He, X., Liaw, B.Y., Santhanagopalan, S., Li, X., Liu, P., Lu, L., et al. (2020). A reliable approach of differentiating discrete sampled-data for battery diagnosis. eTransportation 3, 100051.

29. Liebhart, B., Komsiyska, L., and Endisch, C. (2020). Passive impedance spectroscopy for monitoring lithium-ion battery cells during vehicle operation. J. Power Sources 449, 227297.

30. Shafiei Sabet, P., and Sauer, D.U. (2019). Separation of predominant processes in electrochemical impedance spectra of lithium-ion batteries with nickel-manganese-cobalt cathodes. J. Power Sources 425, 121–129.

31. Ciucci, F. (2019). Modeling electrochemical impedance spectroscopy. Curr. Opin. Electrochem. 13, 132–139.

32. Naha, A., Khandelwal, A., Agarwal, S., Tagade, P., Hariharan, K.S., Kaushik, A., Yadu, A., Kolake, S.M., Han, S., and Oh, B. (2020). Internal short circuit detection in Li-ion batteries using supervised machine learning. Sci. Rep. 10, 1301.

33. Louli, A.J., Li, J., Trussler, S., Fell, C.R., and Dahn, J.R. (2017). Volume, pressure and thickness evolution of Li-Ion pouch cells with silicon-composite negative electrodes. J. Electrochem. Soc. 164, A2689–A2696.

34. Louli, A.J., Ellis, L.D., and Dahn, J.R. (2019). Operando pressure measurements reveal solid electrolyte interphase growth to rank Li-Ion cell performance. Joule 3, 745–761.

35. Barai, A., Tangirala, R., Uddin, K., Chevalier, J., Guo, Y., McGordon, A., and Jennings, P. (2017). The effect of external compressive loads on the cycle lifetime of lithium-ion pouch cells. J. Energy Storage 13, 211–219.

36. Davies, G., Knehr, K.W., Van Tassell, B., Hodson, T., Biswas, S., Hsieh, A.G., and Steingart, D.A. (2017). State of charge and state of health estimation using electrochemical acoustic time of flight analysis. J. Electrochem. Soc. 164, A2746–A2755.

37. Knehr, K.W., Hodson, T., Bommier, C., Davies, G., Kim, A., and Steingart, D.A. (2018). Understanding full-cell evolution and non-chemical electrode crosstalk of Li-Ion batteries. Joule 2, 1146–1159.

38. Feng, X., He, X., Lu, L., and Ouyang, M. (2018). Analysis on the fault features for internal short circuit detection using an electrochemical-thermal coupled model. J. Electrochem. Soc. 165, A155–A167.

39. Zhu, S., Han, J., An, H.-Y., Pan, T.-S., Wei, Y.-M., Song, W.-L., Chen, H.-S., and Fang, D. (2020). A novel embedded method for in-situ measuring internal multi-point temperatures of lithium ion batteries. J. Power Sources 456, 227981.

40. Feng, X., Weng, C., Ouyang, M., and Sun, J. (2016). Online internal short circuit detection for a large format lithium ion battery. Appl. Energy 161, 168–180.

41. Schuster, E., Ziebert, C., Melcher, A., Rohde, M., and Seifert, H.J. (2015). Thermal behavior and electrochemical heat generation in a commercial 40 Ah lithium ion pouch cell. J. Power Sources 286, 580–589.

42. Robinson, J.B., Engebretsen, E., Finegan, D.P., Darr, J., Hinds, G., Shearing, P.R., and Brett, D.J.L. (2015). Detection of internal defects in lithium-ion batteries using lock-in thermography. ECS Electrochem. Lett. 4, A106–A109.

43. Jia, Z., Liu, Z., Vong, C., Pecht, M., and Rotating, A. (2019). A rotating machinery fault diagnosis method based on feature learning of thermal images. IEEE Access 7, 12348–12359.

44. Abada, S., Marlair, G., Lecocq, A., Petit, M., Sauvant-Moynot, V., and Huet, F. (2016). Safety focused modeling of lithium-ion batteries: a review. J. Power Sources 306, 178–192.

45. Chemali, E., Kollmeyer, P.J., Preindl, M., and Emadi, A. (2018). State-of-charge estimation of Li-ion batteries using deep neural networks: a machine learning approach. J. Power Sources 400, 242–255.

46. Finegan, D.P., Darst, J., Walker, W., Li, Q., Yang, C., Jervis, R., Heenan, T.M.M., Hack, J., Thomas, J.C., Rack, A., et al. (2019). Modelling and experiments to identify high-risk failure scenarios for testing the safety of lithium-ion cells. J. Power Sources 417, 29–41.

47. Li, S., He, H., and Li, J. (2019). A cloud-based aging considered vehicle-mounted lithium-ion battery management method: a big data perspective. In 3rd Joint International Conference on Energy, Ecology and

Environment and Electrical Intelligent Vehicles, pp. 181–184.

48. Zhu, J., Wierzbicki, T., and Li, W. (2018). A review of safety-focused mechanical modeling of commercial lithium-ion batteries. J. Power Sources *378*, 153–168.

49. Feng, X., Ouyang, M., Liu, X., Lu, L., Xia, Y., and He, X. (2018). Thermal runaway mechanism of lithium ion battery for electric vehicles: a review. Energy Storage Mater. *10*, 246–267.

50. Yang, C., Finegan, D., and Keyser, M.A. (2019). Integrated multiphysics modeling for improving Li-Ion battery pack safety (36th Annual International Battery Seminar & Exhibit). https://www.osti.gov/biblio/1558359.

51. Zhang, J., Lv, D., and Simeone, A. (2020). Artificial neural network based multisensor monitoring system for collision damage assessment of lithium-ion battery cells. Energy Technol. *8*, 2000031.

52. Wang, B., Zhang, G., Wang, H., Xuan, J., and Jiao, K. (2020). Multi-physics-resolved digital twin of proton exchange membrane fuel cells with a data-driven surrogate model. Energy and AI *1*, 100004.

53. Zhang, Y., Tang, Q., Zhang, Y., Wang, J., Stimming, U., and Lee, A.A. (2020). Identifying degradation patterns of lithium ion batteries from impedance spectroscopy using machine learning. Nat. Commun. *11*, 1706.

54. Krewer, U., Röder, F., Harinath, E., Braatz, R.D., Bedürftig, B., and Findeisen, R. (2018). Review—dynamic models of Li-Ion batteries for diagnosis and operation: a review and perspective. J. Electrochem. Soc. *165*, A3656–A3673.

55. Chen, Z., Andrejevic, N., Smidt, T., Ding, Z., Chi, Y.-T., Nguyen, Q.T., Alatas, A., Kong, J., and Li, M. (2020). Direct prediction of phonon density of states with Euclidean neural network. arXiv, arXiv:2009.05163.

56. Petretto, G., Dwaraknath, S., P C Miranda, H., Winston, D., Giantomassi, M., van Setten, M.J., Gonze, X., Persson, K.A., Hautier, G., and Rignanese, G.M. (2018). High-throughput density-functional perturbation theory phonons for inorganic materials. Sci. Data *5*, 180065.

57. Chen, C., Zuo, Y., Ye, W., Li, X., Deng, Z., and Ong, S.P. (2020). A critical review of machine learning of energy materials. Adv. Energy Mater. *10*, 1903242.

58. Raissi, M., Perdikaris, P., and Karniadakis, G.E. (2017). Physics informed deep learning (part i): data-driven solutions of nonlinear partial differential equations. arXiv, arXiv:1711.10561.

59. Raissi, M., Perdikaris, P., and Karniadakis, G.E. (2017). Physics informed deep learning (Part II): data-driven discovery of nonlinear partial differential equations. arXiv, arXiv:1711.10566.

60. Zhang, L., Lin, D., Wang, H., Car, R., and E, W. (2019). Active learning of uniformly accurate interatomic potentials for materials simulation. Phys. Rev. Mater. *3*, 023804.

61. Zhang, L., Han, J., Wang, H., Saidi, W., Car, R., and Weinan, E. (2018). End-to-end symmetry preserving inter-atomic potential energy model for finite and extended systems. arXiv, arXiv:1805.09003.

62. Lu, L., Meng, X., Mao, Z., and Karniadakis, G. (2019). DeepXDE: a deep learning library for solving differential equations. arXiv, arXiv:1907.04502.

63. Zhao, H., Storey, B.D., Braatz, R.D., and Bazant, M.Z. (2020). Learning the physics of pattern formation from images. Phys. Rev. Lett. *124*, 060201.

64. Wang, Z., and Zhang, Z. (2020). A mesh-free method for interface problems using the deep learning approach. J. Comp. Phys. *400*, 108963.

65. Sirignano, J., and Spiliopoulos, K. (2018). DGM: a deep learning algorithm for solving partial differential equations. J. Comp. Phys. *375*, 1339–1364.

66. Li, W., Bazant, M.Z., and Zhu, J. (2020). A physics-guided neural network framework for elastic plates: comparison of governing equations-based and energy-based approaches. arXiv, arXiv:2010.06050.

67. Bazant, M.Z. (2013). Theory of chemical kinetics and charge transfer based on nonequilibrium thermodynamics. Acc. Chem. Res. *46*, 1144–1160.

68. Lim, J., Li, Y., Alsem, D.H., So, H., Lee, S.C., Bai, P., Cogswell, D.A., Liu, X., Jin, N., Yu, Y.-S., et al. (2016). Origin and hysteresis of lithium compositional spatiodynamics within battery primary particles. Science *353*, 566–571.

69. Thomas-Alyea, K.E., Jung, C., Smith, R.B., and Bazant, M.Z. (2017). In situ observation and mathematical modeling of lithium distribution within graphite. J. Electrochem. Soc. *164*, E3063–E3072.

70. Huang, W., Attia, P.M., Wang, H., Renfrew, S.E., Jin, N., Das, S., Zhang, Z., Boyle, D.T., Li, Y., Bazant, M.Z., et al. (2019). Evolution of the solid–electrolyte interphase on carbonaceous anodes visualized by atomic-resolution cryogenic electron microscopy. Nano Lett. *19*, 5140–5148.

71. Das, S., Attia, P.M., Chueh, W.C., and Bazant, M.Z. (2019). Electrochemical kinetics of SEI growth on carbon black: part II. Modeling. J. Electrochem. Soc. *166*, E107–E118.

72. Smith, R.B., and Bazant, M.Z. (2017). Multiphase porous electrode theory. J. Electrochem. Soc. *164*, E3291–E3310.

73. Han, J., Jentzen, A., and E, W. (2018). Solving high-dimensional partial differential equations using deep learning. Proc. Natl. Acad. Sci. USA *115*, 8505–8510.

74. Qian, E., Kramer, B., Peherstorfer, B., and Willcox, K. (2020). Lift & Learn: physics-informed machine learning for large-scale nonlinear dynamical systems. Phys. D: Nonlinear Phenom. *406*, 132401.

75. Hwasser, M., Kragic, D., and Antonova, R. (2020). Variational auto-regularized alignment for Sim-to-real control. IEEE International Conference on Robotics and Automation (ICRA), 2732–2738.