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# Implications of the BATTERY 2030+ AI-Assisted Toolkit on Future Low-TRL Battery Discoveries and Chemistries

Arghya Bhowmik, Maitane Berecibar, Montse Casas-Cabanas, Gabor Csanyi, Robert Dominko, Kersti Hermansson, M. Rosa Palacin, Helge S. Stein, and Tejs Vegge\*

**BATTERY 2030+** targets the development of a chemistry neutral platform for accelerating the development of new sustainable high-performance batteries. Here, a description is given of how the AI-assisted toolkits and methodologies developed in BATTERY 2030+ can be transferred and applied to representative examples of future battery chemistries, materials, and concepts. This perspective highlights some of the main scientific and technological challenges facing emerging low-technology readiness level (TRL) battery chemistries and concepts, and specifically how the AI-assisted toolkit developed within BIG-MAP and other BATTERY 2030+ projects can be applied to resolve these. The methodological perspectives and challenges in areas like predictive long time- and length-scale simulations of multi-species systems, dynamic processes at battery interfaces, deep learned multi-scaling and explainable AI, as well as AI-assisted materials characterization, self-driving labs, closed-loop optimization, and AI for advanced sensing and self-healing are introduced. A description is given of tools and modules can be transferred to be applied to a select set of emerging low-TRL battery chemistries and concepts covering multivalent anodes, metal-sulfur/oxygen systems, non-crystalline, nano-structured and disordered systems, organic battery materials, and bulk vs. interface-limited batteries.

## 1. Introduction


Fast-tracking the discovery and optimization of new battery concepts, materials, and cell designs requires more than obtaining a detailed understanding of the underlying physicochemical processes that govern the performance and durability of the batteries. It critically depends on the development of versatile toolkits and modular platforms that can assist and complement the researchers and scientists in performing the necessary investigations and developments at a much faster pace. Many of these tools rely upon advances in complementary scientific disciplines that can be adapted to the specific needs and challenges in the chemistry and materials science of batteries. Historically, one of the most significant driving forces for accelerated research in materials science has been high-throughput experimentation

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and simulations. Originating in the 1950s, for example, in the fields of biology with the use of multi-well microtiter plates<sup>[1]</sup> and metallurgy where scientists sought to discover new dental alloys,<sup>[2]</sup> comprehensive platforms are now being developed for the advancement of autonomous research.<sup>[3]</sup> The recent flourish in machine learning (ML) for materials and energy science, and methods for AI-orchestrated materials discovery, may likely provide a similar or even larger boost.<sup>[3–5]</sup>

While highly specialized methods, experimental rigs, and testing equipment can be developed and optimized to solve a specific scientific challenge such as unraveling the solid-electrolyte interphase (SEI) in Li-ion batteries (see discussion in Section 11), they often require substantial and time-consuming modifications and adaption to be applicable to new battery chemistries, materials structures or cell designs. As a multitude of different battery concepts and cell designs are currently under investigation as potential candidates for the elusive ultra-performance, durable, safe, and sustainable batteries of the future, it begs the question: how can the methodologies and toolkits developed in BATTERY 2030+ and related large-scale initiatives quickly be adapted and applied to accelerate the discovery and development of new low-technology readiness level (TRL) battery chemistries and concepts?<sup>[6,7]</sup> In this manuscript, the main focus is on the importance of ensuring generalizability and externalizability of the developed toolkits, such as software codes, apps, REST application programming interfaces, autonomous workflows, AI-based methods, data interoperability, as well as modularity and versatility of the experimental and computational setups.

While a plethora of great reviews about the general acceleration of materials science has emerged,<sup>[8–10]</sup> this perspective aims at showcasing how the AI-assisted toolkits developed in BATTERY 2030+ and related large-scale research initiatives to accelerate the development of Li-ion batteries can be transferred and applied to meet these challenges. We highlight this on five representative cases for emerging low-TRL battery chemistries and concepts: I) Multivalent anodes, II) Metal-sulfur/oxygen batteries, III) Non-crystalline, nano-structured and disordered systems, IV) Bulk vs. interface-limited batteries, and V) Organic battery materials.

We first identify the main scientific and technological bottlenecks that need to be resolved and then outline the applicability of the AI-assisted toolkit with a specific focus on transferability and modularity. As the detailed technical nature of critical elements in the BATTERY 2030+ infrastructure (like autonomous workflows, operando characterization, and the battery interface ontology (BattINFO [<https://github.com/BIG-MAP/BattINFO>]) developed in the Battery Interface Genome—Materials Acceleration Platform (BIG-MAP) project, are described in other papers in the special issue, we focus mainly on the implementation in the five low-TRL examples.

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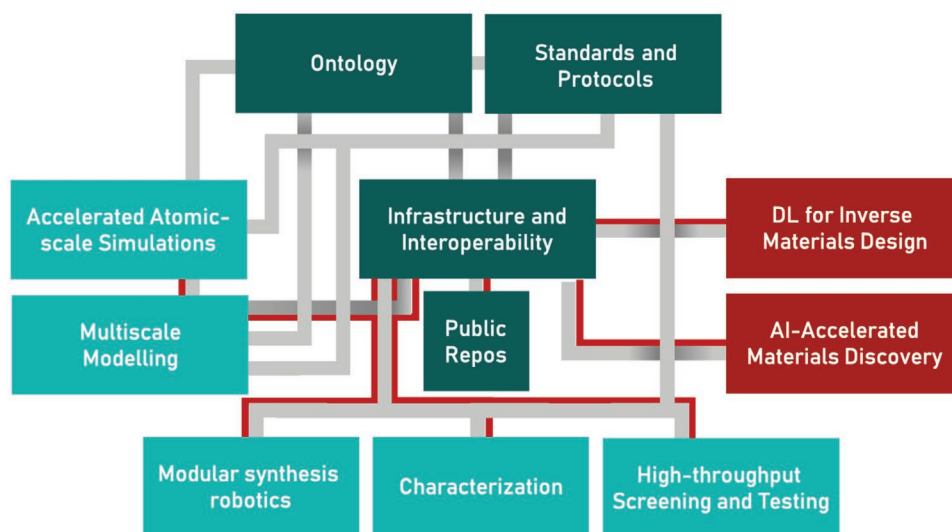
## 2. Shared Data-Infrastructures, Modularity, and Transferability

Putting battery laboratory researchers in a position to dedicate their focus entirely to the interesting science and innovation instead of being burdened by tedious, repetitive tasks in the laboratory or at their computer calls for a transformative approach to the innovation cycle. The general idea is to deploy highly efficient closed-loop technologies,<sup>[6]</sup> digitalization, automation, and autonomous integrated experimental and computational workflows to leave human scientists to deal with the main R&D challenges.

Discovery and optimization of energy materials and interfaces is a tough challenge, which historically has to some extent relied on serendipity.<sup>[11]</sup> Examples range from the discovery of antibiotics and vulcanization of rubber to the discovery of shape memory alloys and modern battery materials. Often these materials were discovered with unusually high functional properties or with, at the time, unknown combinations of functional properties. With the ever-growing complexity of materials, interfaces, and their intertwined properties, it is increasingly hard to discover new and improved materials as specific functional properties appear mutually exclusive, for example, ductility and strength. However, optimizing materials is challenging as trends exist only in high-dimensional spaces and appear mostly at delta-like phase boundaries, making gradient descent futile. Such closed-loop implementations can also help eliminate potential anthropogenic biases in the training data<sup>[12]</sup> and procedures,<sup>[13]</sup> and optimize the exploitation and exploration of computational and experimental space.

A common theme revolves around integrating the tools of a platform and its assets like data and data management.<sup>[14]</sup> Materials Acceleration Platforms<sup>[3,15]</sup> integrate these elements that can lead to a greater pace in research. A foundational building block for the success of any MAP is therefore not “just” making data Findable, Accessible, Interoperable and Re-usable FAIR (Findable, Accessible, Interoperable and Reusable),<sup>[16]</sup> which is a challenge in its own right, but also making the tools transferable and externalizable to the battery community at large, for example, through the BIG-MAP app store (<https://big-map.github.io/big-map-registry/>) and GitHub repository (<https://github.com/BIG-MAP>), in such a way that new data can be acquisition, generated and analyzed on-the-fly.<sup>[17]</sup>

BATTERY 2030+ focuses on developing tools and methods for obtaining the necessary understanding of the spatio-temporal evolution of interfaces and interphases across relevant scales in the most cost-efficient manner. This is done to enable the inverse design of superior battery materials and interfaces in particular, as it is a challenge that transcends all battery chemistries and concepts. In addition to a holistic and FAIR data infrastructure, adhering to specific ontologies such as BattINFO and data schema spanning all parts of the battery discovery value chain, two aspects are critical for the development of a fully autonomous and versatile battery MAP, that is, modularity and transferability to new battery chemistries and concepts (**Figure 1**). The tools developed should ultimately be fully integratable to facilitate experiments by autonomously launching simulations and simulations initiating and running experiments through the BIG-MAP orchestration infrastructure.



**Figure 1.** Data exchange within the BIG-MAP project highlighting the backbone for inverse materials design and AI accelerated materials discovery to be successfully deployed. Core research expertise on the one-off type of method deployments exists across different workgroups. Still, the interconnection through a shared ontology, compliance to standards, and operation on a commonly shared and interoperable data infrastructure are necessary for AI and machine learning methods to be deployed across BIG-MAP. Highlighted are the data exchange focused connections; details are found in Castelli et al.,<sup>[14]</sup> which served as the basis for this Figure.

## 2.1. Transferability

While specific approaches and methods are traditionally developed to solve a particular scientific challenge, say developing a neural network potential capable of describing long time- and length-scale dynamics of a given electrolyte formulation in the presence of positive (negative) electrode and under an applied bias, this tool in itself will not be transferable to other electrolyte compositions or electrode materials, but the workflows themselves needed to develop, train and test the potential utilizing the shared multi-sourced data will be made available to the community through the BIG-MAP app store and GitHub repository.

## 2.2. Modularity

As new low-TRL battery chemistries and concepts are introduced and investigated, new and specialized modules may likely be required, such as specific synthesis techniques (e.g., hydrothermal synthesis) or characterization modules (aqueous organic redox couples), but these modules should be easily exchangeable and integratable in the battery MAP and the underlying data infrastructure (Figure 1).

By focusing on the key scientific and technological challenges in the five low-TRL battery chemistries concepts outlined above, we will illustrate how the AI-based toolkits currently under development in BATTERY 2030+ can be adapted and applied to these cases.

## 3. Predictive Long Time- and Length Scale Simulations of Multi-Species Systems

In recent years, substantial progress has been made for ML potentials capable of describing multiple spatial and temporal

scales of the evolution of reactive liquid-solid/amorphous systems with quantum mechanical (QM) accuracy, describing up to 100 000 atoms for long time-scale simulations.<sup>[18]</sup> Using a Gaussian approximation potential,<sup>[19,20]</sup> molecular-dynamics, Deringer et al., were capable of accessing the time scales needed to predict distinct electronic features that can be compared directly to ultrafast spectroscopic techniques, and the experimentally relevant length scales for the description of (poly-)crystallization in amorphous silicon.<sup>[18]</sup> Extending the first principles approach beyond the electronic structure modeling and retaining QM accuracy for long time- and length scale molecular dynamics (MD) simulations is critical for ensuring the models' predictive capabilities and enabling inverse design of new battery materials and interfaces.

There are particular and general challenges in developing ML potentials for battery materials and interfaces independent of the precise regression scheme used. The development of externalizable tools to deal with these challenges is currently under development in BIG-MAP and plays an essential role in the BATTERY 2030+ Roadmap.<sup>[21]</sup>

Thus far, most ML potentials – that are generally applicable to a wide range of structures and temperature and pressure state points – have been created for systems that contain just a few different chemical elements: elemental compounds,<sup>[18]</sup> metal oxides,<sup>[22]</sup> water,<sup>[23]</sup> etc. However, when modeling battery materials, just the description of the electrolyte necessitates using at least half a dozen elements. This presents a substantial challenge because the two widely used descriptors for building ML models (atom centered symmetry functions<sup>[24]</sup> or smooth overlap of atomic positions<sup>[25]</sup>) scale poorly with the number of elements; in current formulations quadratically. This leads to critical slowdowns when going from elemental materials to 3-4-5 different elements and effectively prevents the construction of good models with 7-8-9 elements, which are often needed for simulations of, for example, realistic battery electrode-electrolyte interface systems. There are efforts toward

simplifying the descriptors for these cases, the goal being to make them scale at most linearly with the number of different elements,<sup>[26,27]</sup> but there is insufficient evidence in the literature as to their overall efficacy for the simulation of complex materials.

The description of liquid organic electrolytes requires that one captures both intra- and intermolecular interactions. Traditionally, force fields would treat these separately during parametrization and add the different contributions to form the complete force field. This construction breaks down if the model describes chemical reactions (e.g., at the electrode-electrolyte interface) because the identity of the molecular constituents changes, and the notional separation into intra- and intermolecular energy contributions are ill-defined during a chemical reaction. There are as yet no ML potentials reported that could describe the basic properties of a molecular liquid other than water (which has relatively strong intermolecular interactions due to the hydrogen bond network), even just in the bulk phase, without any reactions, without explicitly relying on separating energy scales in this way.

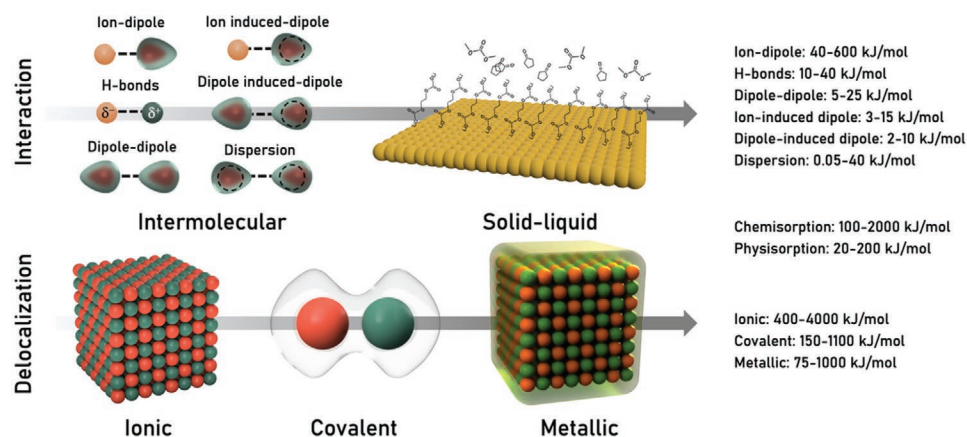
These challenges only become even more complex when dealing with interfacial dynamics, as the encountered forces in the ionic/metallic bonding in the electrode(s) and at the surfaces during the formation of chemical (metal-organic) bonds at the surface are separated further (Figure 2).

#### 4. Toward Inverse Design of Battery Interfaces

Two different phases in contact generally lead to local charge separation. Such polarization effects strongly influence the behavior of the batteries and need to be dealt with when electrode-electrolyte interfaces are modeled. Lithium-ion batteries contain a wide range of scenarios where charge transfer, charge transport, and polarization phenomena play decisive roles. Their modeling is a challenge, as is the experimentation.<sup>[28]</sup> It is well documented in the literature that polarizable force-field models systematically predict higher self-diffusion and ionic conductivity and faster motional

relaxations than the non-polarizable models for many solid and liquid electrolytes, as well as for ionic liquids and ionic electrode materials (see, for example, the review article by Bedrov et al.<sup>[29]</sup> and references therein). Likewise, the preferred coordination numbers and coordination motifs can change drastically when polarization effects are taken into account in the simulation model. Given such findings, it is not surprising that continuous efforts during decades have been devoted to the development of polarisable and advanced force-fields for application in the battery context, as exemplified here by a recent battery-related interface study regarding solvent decomposition reactions at the anode/electrolyte interface using ( $\leq 2.5$  ns) long MD simulations with the ReaxFF polarizable force field.<sup>[30]</sup> The considerable efforts spent in the literature on the development of polarizable interaction models for complex energy materials applications reflects both the great need for efficient force fields that allow large-scale, long-time computer simulations and the formidable challenge that it constitutes to generate force fields that accurately capture the electronic effects.

Another route taken in the literature presents the opposite scenario: small-system, short-time MD simulations where the electrons are still present in the calculation of the forces, so-called *ab initio* MD (AIMD) or DFTMD simulations. One important target area here has been the exploration of the reactivity at the electrode-electrolyte interphase and the growth of the SEI, which consists of insoluble organic and inorganic species that result from the decomposition of solvents and salts from the electrolyte, as redox processes involving both ion and electron transfer are believed to occur. Dozens of AIMD simulations have been published in many cases on the topic of reactivity and electrolyte degradation. Even rather elaborate electrode-electrolyte/additive systems have been explored.<sup>[31]</sup> However, the size of the simulation systems and the time scales are far too small/short for proper statistical sampling of many interface properties and quantities of interest. Typically, the current state of the art is a thin small-area electrode model with a dozen or fewer solvent molecules, and ions studied for some tens of picoseconds. The short time spans reachable are



**Figure 2.** The large variations in inter- and intra-molecular forces for the organic, aqueous, solid, and interfacial systems pose a massive challenge for the development of machine learning potentials for battery interfaces.



particularly limiting because crucial ion transport at the interface and especially in the SEI layer are believed to be very slow processes. The interplay between thermodynamics and kinetics constitutes a major challenge.

As discussed above, proper ML potentials combine the best of the two approaches mentioned, allowing to combine, at reasonable computational costs, a sophisticated description with long observation (i.e., simulation) times. ML protocols for isotropic two-component electrolyte systems are currently under development in BIG-MAP; the future extension to multicomponent solid-liquid interphase/face systems will build on this development, which entails several highly challenging barriers to overcome for these complex systems. One class of such interface-invoked hurdles is long-range anisotropic electrostatic interactions, polarization, and charge transfer, which will occur at the electrode-electrolyte interface and between solid-solid interphases affecting not only reactions but also the atomic/ionic structure and dynamics. In a longer-term perspective (see Roadmap paper by Edström et al. in this issue),<sup>[21]</sup> these effects need to be properly accounted for in the generation of the QM training data and the ML potentials. Furthermore, the different chemical bonding nature of the various compounds and molecules that meet, interact, and react at the interface makes the development of adequate QM methods non-trivial. It necessitates efficient benchmarking protocols, efforts that are ongoing within BIG-MAP.

One of the most significant difficulties added when going from electrolyte modeling to battery interface modeling is the lack of solid knowledge of (most aspects) of the interface structure. Often, not even the composition of these phases is known. The modelers will work in tandem with experimentalists, sharing structural information from different sources. Here, structural characterization will play a key role, and on the modeling side, AI-enhanced automated workflows will be developed to assist in the interpretation of experimental interface spectra.

Yet another major challenge is the representation of the interface under experimental and working conditions, including the coupling to external fields and allowing for charge transport through the battery. Initial promising efforts along these lines are discussed in refs. [32–34].

Beyond the connection of theory and experiment is the interrelation of interface and system-focused experiments. An emblematic example of the challenges and potential solutions for bridging the disconnection between interfaces and systems in the experimental materials sciences can be found in the field of high-throughput electrochemistry. With scanning droplet cell (SDC) electrochemistry,<sup>[35]</sup> a new pulsed charging protocol has been developed for Zn metal deposition. The challenge is that Zn forms dendrites at virtually any charge rate. Using SDC, it was discovered that a specific pulsed charging method could completely prevent dendrite formation upon Zn metal plating.<sup>[35]</sup> The same process was then transferred and used to extend the lifetime of a Li-metal/LiFePO<sub>4</sub> well beyond 6500 cycles at a >1 C charge rate.<sup>[35]</sup> Generally speaking, only some effects observable on the millimeter scale are easily transferrable to complete systems. However, metal intercalation, insertion, or electrode conversion reactions are governed by interfacial processes rather than system effects like thermal management or electrode dimensions. Therefore, many

first-order effects in a system can be observed in model systems dealing with just one interface/interphase. Therefore, surrogate<sup>[36]</sup> or transfer learning models<sup>[37]</sup> alkali as those developed in BIG-MAP, aim to translate from interfacial to system-level effects is necessary.

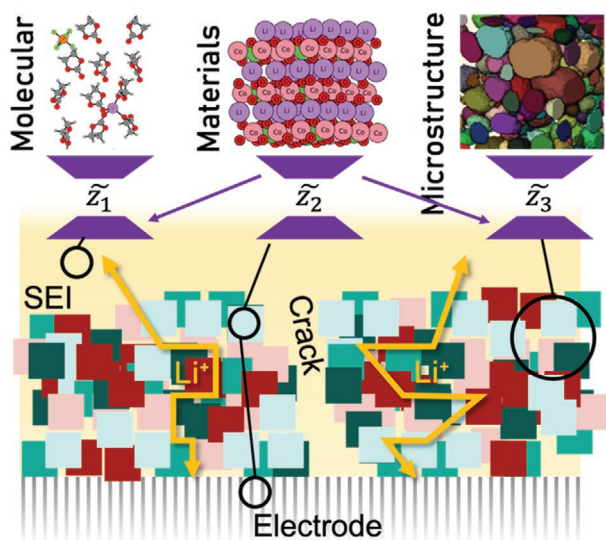
## 5. Deep Learned Multi-Scaling and Explainable AI

### 5.1. Generative Models and Inverse Design

Deep generative models can learn the underlying probability distributions in the input space<sup>[38]</sup> and thus be used to encode physics-driven constraints in materials structures at the atomic<sup>[39]</sup> as well as the continuum scale<sup>[40]</sup> from large, validated datasets. Trained models can then be utilized to create new and novel structures while retaining plausibility as potential battery materials. For a detailed description of the current state of the art, please refer to SOA Fichtner et al. in this issue.<sup>[41]</sup>

While generative models can help create large structure libraries autonomously from smaller ones, screening processes to search for candidates that satisfy property requirements are needed during the design process. Alternatively, true inverse design can be achieved with conditional generative models that learn the probability distribution in structural space conditioned to the properties.<sup>[42]</sup> Thus, structures can be probabilistically generated for target property value. With appropriate training structure-property datasets, conditional variational autoencoders<sup>[43–45]</sup> and conditional generative adversarial network<sup>[46,47]</sup> can be trained for this purpose. Additionally, simulation environments that provide property estimations and structural validity can be used for training reinforcement learning (RL) agents for targeted structure generation.<sup>[48,49]</sup> In principle, target-property conditioned structure generation can be done for multiple properties at once, allowing Pareto-style inverse design for multiple target properties. Nonetheless, it is very data-intensive to learn distributions across multiple properties unless the properties are inter-correlated.

While generative models including conditional variants for molecules and bulk solids have seen initial success, batteries are interfacial systems incorporating numerous liquid and condensed matter phases. The observed properties are controlled by structures at multiple length scales<sup>[50,51]</sup> and their evolution over time. Thus, the inverse design of high-performance batteries needs a new class of generative models that are universal to solid/liquid and organic/inorganic bonds and phases and work with multiscale structural representations and properties. In BIG-MAP, we investigate a possible path forward using hierarchical VAEs<sup>[52,53]</sup> that work with multiple latent spaces for multiple length scales conditioned to each other (Figure 3).<sup>[54]</sup> As it learns structural correlations across time and length scales from large datasets that describe battery electrochemical interfaces and properties, such a multiscale generative model can also perform scale bridging through one-shot generation of multiscale structures without the need for step-by-step multi-scaling. It is expected that many other and complementary ideas will need to be envisaged and pursued, given how challenging the target is.



**Figure 3.** Utilize embedding models (denoted by purple prisms) to interrelate the different phenomena and observations on the molecular, materials, and microscale. These models are used to generate latent space embeddings that help in the understanding of how, for example, processes at the molecular level influence the microscale. Exemplarily shown is an SEI on an electrode highlighting the diversity of the SEI composition and the complex ion diffusion through it.

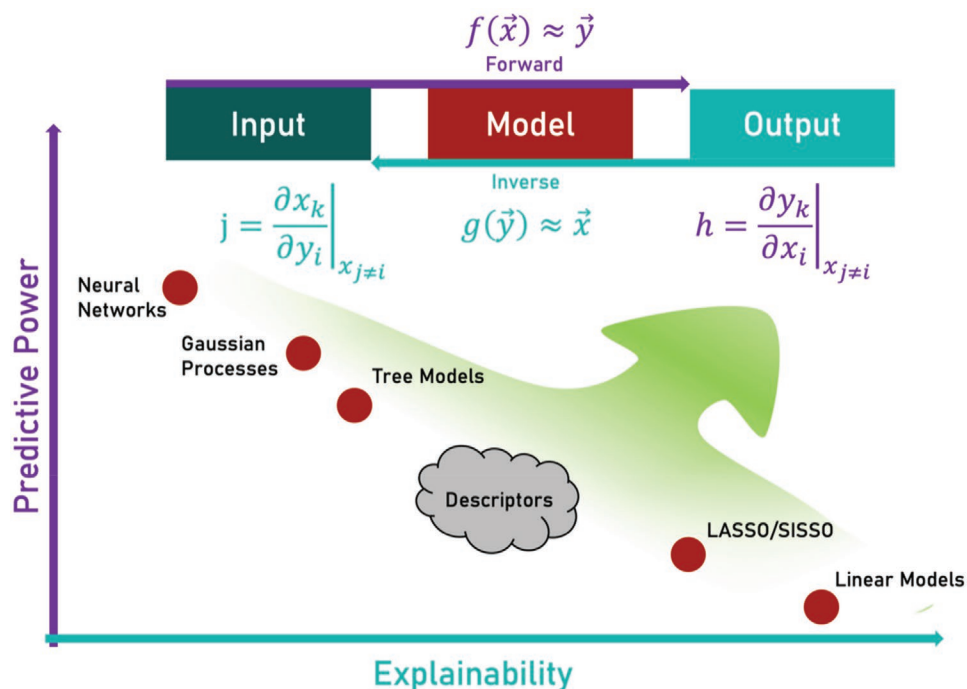
## 5.2. Explainable AI and Descriptors

Deep learning models are increasingly utilized as a key tool in data-driven materials modeling in battery materials and

interfaces. The success of deep learning emanates from the capacity to model very complex functions of input features. However, the inherent black-box nature stops researchers from understanding what functions of features are utilized to arrive at the output. Predictions from deep models must be driven by combinations of data features appropriate in the context of the real underlying physical phenomena under investigation. Interpretability is crucial to achieving a physics-driven approach to deep learning in electrochemical sciences.

‘Explainable AI (XAI)’<sup>[55,56]</sup> provides a path to open the black box models such that data patterns can be connected to the model outputs; it can open up a new pathway toward utilization of big data in scientific discovery via data-driven realization of chemical laws and design principles, for example, by moving away from simple linear descriptors<sup>[57]</sup> to complex ones combining multiple features<sup>[58]</sup> (Figure 4). A detailed description of the current state of the art in XAI is given in ref. [41].

Structure-property trends derived from such descriptors are easy to reason and verify, and they provide the physics-driven grounding and credibility to the developed models. As understanding complex multi-scale inter-correlated phenomena at the battery interface and other such systems are limited, ML models in electrochemical sciences have traditionally relied on comprehensible models with moderate accuracy.<sup>[59,60]</sup> However, there is a clear trend in deep learning models quickly gaining popularity in this application space. This highlights the conflict between predictive accuracy and explainability while building ML models. Often, the highest performing methods (e.g., deep neural networks) are the least explainable, and the most explainable (e.g., linear regression) have low accuracy. The goal of the developed XAI models is to make deep models



**Figure 4.** Explanation of forwarding and inverse predictions. Common AI methods in materials science work in forward prediction, but few can inversely predict input parameters. A proxy could be Explainable AI that effectively allows studying the partial (high dimensional and sparse) derivative of the output by the input.

intelligible to humans by explaining how the output is formed from what kind of salient features are in the input variable. Explainable AI is also “reliable AI,” as the predictions come from the correct reasoning. An alternative way to interpret and explain black-box ML models is to design inherently interpretable models without significant loss of accuracy.<sup>[61]</sup>

XAI methods are broadly categorized into four groups—perturbation-based, function-based, sampling-based, and structure-based. Perturbation-based methods study changes in output due to perturbation or masking in input.<sup>[62,63]</sup> The function-based approach does functional analysis on the deep model like sensitivity analysis, Taylor decomposition approximation, and gradient analysis.<sup>[64,65]</sup> Sampling-based methods try to approximate the prediction pattern locally with surrogates to make local explanations in the manifold.<sup>[66,67]</sup> Finally, structure-based XAI relies on analyzing the structure of the model through decomposition into functional components to be explained independently, such that the decision function as a whole can be understood.<sup>[68,69]</sup> Most of the XAI methods have been developed for pixel-based models and are thus most suitable for meso- to macro-scale data from battery systems. However, recently XAI methods for graph neural networks have been developed.<sup>[70–72]</sup> Identifying substructures both in data and in the model gives the possibility of understanding the structural patterns leading to specific properties and the governing equations that are discoverable. Thus, one can define an infinite design space in local regions with fast surrogates and utilize agent-based methodologies for multiscale design.

### 5.3. AI-Assisted Materials Characterization

While there is a substantial focus on developing machine/deep learning models aiming at predicting new materials with better functional properties as described above, there is a significant research direction of using AI-based models for automated/autonomous materials data analysis. Namely, there has been an over 15 year-long development of tools for the automatic phase mapping problem in combinatorial materials science. Based on spectral decomposition,<sup>[73]</sup> there has been tremendous<sup>[74–76]</sup>

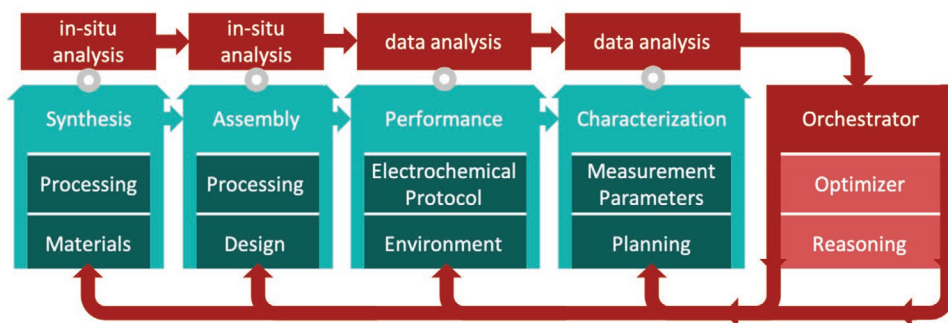
progress for phase mapping with, for example, the Gibbs’ Phase Rule as a constraint. Moreover, it is even possible to extract optimized crystal structures with physically correct orientation and phase amounts from high-throughput X-ray diffraction.<sup>[77]</sup>

Prior to applying any machine learning approach, there is typically much data curation needed like noise filtering or background subtraction, with the latter one recently being demonstrated with algorithms that consider the entire dataset taken going beyond single spectra.<sup>[78]</sup> Even seemingly simple tools allowing users to perform reliable peak identification can accelerate the speed at which the research can be conducted. AI-assisted tools developed in BIG-MAP for these tasks will therefore be bundled in servers that offer scientists reliable and traceable data analysis tools developed for batteries and beyond accelerating research beyond the lifetime of the project.

## 6. Self-Driving Labs, Closed-Loop Optimization, and Discovery

The six scientific projects currently active under the umbrella of BATTERY 2030+ strive to build AI-assisted tools and platforms to position scientists for a new level of cooperation, where the physical and infrastructural boundaries between research groups and institutions are diminishing. Viewed from a top-down perspective, such interactions can be seen as the tight interconnection of different research groups and the accelerated research tasks<sup>[10]</sup> therein. The seamless integration of research tasks automatically triggering the next pertinent task is then considered so-called closed-loop<sup>[79,80]</sup> or, on a larger scale, a self-driving lab<sup>[81–86]</sup> when fully interconnected, even a materials acceleration platform. Similar to the levels of autonomous driving, Stein and Gregoire<sup>[10]</sup> tried to assess different early-stage closed-loop discovery cycles in chemistry.<sup>[6,80]</sup> This assessment is based on the number of (accelerated) tasks, their element research task with the lowest throughput, the total number of unattended runs, the level of materials intelligence,<sup>[87]</sup> and the automation complexity of the closed-loop.

As shown in **Figure 5**, there is a need to exchange data, samples, and analyses. Within an initiative like BATTERY 2030+,



**Figure 5.** Schematic flow of data (red) and physical samples or simulation data (turquoise) in an exemplary closed-loop optimization and discovery loop. Building a secondary battery involves several synthesis and processing steps, which need to be planned and optimized. After synthesizing materials, for example, an active electrode material, the battery must be assembled into the appropriate form. Here design and processing, that is, how the battery is built, is an input. Subsequently, the manufactured battery is analyzed for performance, and further characterization of the materials, for example, after cycling, can be performed. Crucial to the tight integration of these steps is the capability of autonomous inline analysis of acquired data. Presently, a focus upon optimizers and reasoners is placed. However, the AI enhanced data analysis, for example, for analyzing X-ray diffraction data, yields fascinating science and is a prerequisite for self-driving labs.



the collaborations between multiple groups, therefore, need to exceed regular scientific exchange of ideas, data, or samples and allow the participating partners to deploy or initiate the running of an experiment, simulation, or data retrieval at the given infrastructure providing the most valuable data at the lowest cost (e.g., in terms of time or cost).<sup>[14]</sup> At the level of the individual research groups, there is a critical need to accelerate and integrate the different research instruments available, carefully aligning the integration, acceleration, and automation in interconnected laboratory instruments.<sup>[88]</sup> Core to these interconnections are protocols that allow machines to communicate with each other, with a recent trend to implement these communications as web servers using standard web requests or more advanced OPCUA<sup>[89]</sup> servers that even allow requests for “proper” formulation. Proper, in this context, refers to ontology-aware requests and answers. Gathered data then needs to be stored in a form that goes beyond the FAIR (Findable, Accessible, Interoperable and Re-usable) data principles.<sup>[16]</sup> meaning that given the (meta) data, scientists in other labs are poised to rerun an experiment or simulation without much overhead, given they have access to the proper equipment/infrastructure. Some tools are already deployed, for example, the metadata extraction module,<sup>[90]</sup> and these will be further developed within BIG-MAP to strive to make the generation of simulation and laboratory workflows a facile task. Examples include the Materials Cloud<sup>[91–93]</sup> and Simstack for computational workflows and ChemMotion<sup>[94]</sup> for organic chemistry-focused research, and KADI4Mat<sup>[95]</sup> as an institution-wide deployed data management platform for materials science. Other examples include MEAD<sup>[96,97]</sup> and HTEM<sup>[98]</sup> for the experimental sciences showcasing the combined community efforts to develop interoperable data repositories. However, self-driving labs<sup>[81]</sup> require more than high-level data management, that is, the capabilities of orchestrating the machines.<sup>[88]</sup> Orchestration in a laboratory context means that there are interfaces for different hardware, automated data analysis, and autonomous planning<sup>[99]</sup> of the next Pareto optimal<sup>[100]</sup> experiment(s) using domain-specific optimizers.<sup>[101]</sup> A strong focus is placed in BATTERY 2030+ on ensuring the transition from automated to autonomous materials discovery.

Given the community adoption of these data management or even workflow tools, hardware and software interconnection remains the most significant remaining challenge. While commercial frameworks like LabView exist, scientists developed tailored solutions like ChemOS,<sup>[102]</sup> ESCALATE,<sup>[103]</sup> and the Chemputer.<sup>[83,104]</sup> Within BIG-MAP, there is a strong push to integrate closed-loop capable workflows in the simulation and experimental realm. To this end, not just transfer learning across the approaches is necessary, but also a new level of cooperation between experimental and computational groups is necessary. Within BIG-MAP, these collaborations are fostered through the construction of work packages (Figure 1) that distribute work across the disciplines equally and through a publicly available app store.

## 7. AI for Advanced Sensing and Self-Healing

The rapidly increasing demand for batteries needs to be complemented with improved quality, reliability, safety, and prolonged

lifetime.<sup>[105–107]</sup> This can be realized with a preventive or curative approach. Cell components and their interfaces are based on highly reactive chemistries, which result in copious parasitic reactions compromising quality, reliability, safety, and lifetime aspects under certain internal or external conditions. The use of preventive steps with properly designed interfaces and interphases can enable improved properties, albeit preventive measures cannot completely stop chemical or physical damages compromising the acceptability and safety of the battery cells. Moreover, the battery cells are operating under different conditions, and their chemical and electrochemical reactions, as well as the physical and mechanical processes occurring during the multiple charge-discharge cycles, contribute to the overall battery aging that determines the lifetime of the battery cells, their safety issues, and thus their overall quality and reliability. Processes like the disintegration of electrode composites, pulverization of active particles, and irreversible reactions producing different degradation products, for example, transition metal ions and organic species dissolved in the electrolyte, continuously occur during operation under other conditions. The released metal ions and organic species can diffuse or migrate through the separator and deposit at the anode surface or trigger shuttling self-discharge mechanisms. Apart from that, continuous aging occurs mainly due to the thickening of the SEI layer, which introduces additional resistance, consequently influencing the performance and safety. Degradation of inactive components, such as corrosion of current collectors, degradation of binder, decomposition of conductive additives, and loss of integration in the separator, should be considered in the engineering step of battery cells.<sup>[108–111]</sup>

The detection of irreversible changes is a first step toward obtaining better reliability and increasing the lifetime and consequently the overall quality of the battery. Hence, accurate states must be obtained to prevent extra degradation of the cells, inefficient operation outside the safe-operation area, or worst-case scenarios of thermal runaways. These are categorized into state-of-charge (SoC),<sup>[112]</sup> state-of-health (SoH)<sup>[113]</sup> in terms of capacity and power, and state-of-safety. In contrast, a general term known as State-of-X (SoX) describes all the accounted state estimation processes. State-of-the-art multi-sensory approaches are now under research in several BATTERY 2030+ projects, for example, to enlarge the battery's lifetime, that is, temperature sensors, acoustic sensors, optical fibers, gauge sensors, electrochemical sensors, etc.

In recent studies, the possibilities of utilizing ultrasonic acoustic sensors<sup>[114]</sup> and non-destructive testing probes with high frequency<sup>[115]</sup> are explored for SoX estimation.<sup>[116]</sup> Also, the mechanical properties of the cells, such as the internal pressure, compression, or strain in the battery cell, can be monitored by tracking the volume changes and utilized for both SoC and SoH estimations.<sup>[117]</sup> Optical fiber sensors have been able to track chemical events such as SEI formation and structural evolution, which can correlate to SoH estimation and detect cell temperature gradients.<sup>[118]</sup> Additionally, thermal sensors and electrochemical impedance spectroscopy (EIS) can play a role in developing non-invasive state estimations.<sup>[119]</sup> Furthermore, the smart combination of multiple sensors may be even more beneficial for enhanced tracking of the battery's health, lifetime, and failure detection. However, for the successful development

of any SoX estimation algorithm, it is essential to understand the signals and parameters measured by each sensor and to correlate those sensor values accurately to the degradation mechanisms that might be happening in the battery cell.

The development of smart and multi-sensory functionalities incorporated within the battery cells should be linked with external logical operators, which will determine the level of damage in the cell and perform an appropriate action. Nowadays, the SoX is rarely performed online, as it is challenging to implement. Nevertheless, with the developments in machine learning and on-demand cloud computing, the capabilities of the BMSs can be significantly increased.<sup>[120]</sup> Different scenarios can be envisaged. In the most advanced scenario, signals obtained with sensors should adhere to the relevant battery ontology, such that the data can be utilized autonomously by the models to predict if/when to trigger the stimulus of the self-healing process in the battery cell.<sup>[4]</sup>

Nature is full of examples where self-healing occurs spontaneously and enables the prolonged lifetime of most living organisms.<sup>[121–123]</sup> Self-healing functionalities in nature have been developed through evolutionary processes. Processes evolved over millions of years today inspire scientists to use known approaches in different technologies. This type of innovation is called biomimetic design and has inspired many of our best creations. Additional self-healing functionalities have been developed with the occurrence of medicine and with the vectorization of medicaments. Self-healing functionalities developed in nature can be considered autonomous/spontaneous approaches in prolonging lifetime.

In contrast, approaches developed in medicine are traditionally related to the non-autonomous/on-demand self-healing functionalities. Applying these self-healing approaches to battery systems will be essential to create additional functionalities for different parts of the battery cell, like the composite electrodes and separators, as developed self-healing functionalities should have preventive or curative functions. While first serving as prevention of contamination of one electrode with degradation products from the other electrode or preventing the loss of integrity and homogeneity of the composite electrode, the latter should focus on on-demand administration of molecules that can impact quality, reliability, safety, and lifetime of cells.<sup>[124]</sup>

## 8. Multivalent Anodes

Rechargeable battery technologies relying on multivalent metal negative electrodes (Ca, Mg, or Al) hold the promise of sustainability and high energy density and therefore have been intensively studied in recent years.<sup>[125–131]</sup> Their practical deployment is, however, plagued by a number of bottlenecks, including not only issues related to the migration of multivalent charge carriers within the electrolyte and the positive electrode but also, and most importantly, aspects related to the negative metal electrode.

Electrochemical plating/stripping of these metals can be performed in non-aqueous electrolytes and, depending on the electrolytes stability window, can compete with electrolyte reduction on the fresh surface of the metals. Although some electrolytes show high stability at the voltage of metal

deposition, their oxidative stability is then compromised. They are thus compatible only with positive electrode materials operating at low potential. Hence, the energy density for the full cell is less attractive. A higher energy density can be obtained with high voltage oxides, sulfur, or redox-active organics that require careful selection of solvents and salts with at least quasi thermodynamic stability in the voltage window of their operation.<sup>[126]</sup> While the rechargeability (i.e., reversibility of the plating/stripping process) may seem a simple matter, at first sight, smooth plating of metal is well known to be a complex issue from classical electrochemistry. Besides the influence of a possible surface passivation layer on the electrodes (see below), the location of the electrodeposit may not be the same specific place where the stripping takes place due to issues related to the current distribution, fluid dynamics, and crystal growth.<sup>[132,133]</sup> All the mentioned processes involve a particular polarization which is also most affected by the desolvation of multivalent cations from electrolyte solvent molecules. The energy required for desolvation corresponds to several hundredths of millivolts, which may be enough to make the process of plating competitive to the electrolyte degradation on the fresh electrode surface.<sup>[134,135]</sup> The electroplating process thus typically results in uneven deposits, which in the most severe formation can be represented as dendrites.<sup>[132,136]</sup> Metal dendrite formation upon cycling was early identified as an impeding factor for developing batteries based on lithium metal negative electrodes, as they may penetrate the separator and result in a short-circuit causing safety risks. Electrodeposition of dendrites results from a diffusion-limited process on the crystal facets, the deposition potential being significantly shifted negative from the equilibrium potential of the  $M^{n+}/M$  couple. Under such conditions, an anisotropic growth is highly favored by the minimal surface diffusion of the adsorbed cations before the charge transfer occurs,<sup>[133]</sup> and an improved hemispherical diffusion, therefore, facilitates the dendrite formation at the tip of the dendrite.<sup>[137]</sup> The current distribution associated with the cell geometry and cycling conditions (negative/positive ratio, particles size, shape, C rate, temperature, etc.) and  $M^{n+}$  diffusion within the electrolyte are key parameters controlling the formation of uneven deposits.

An important parameter affecting the plating/stripping process is the nature of the electrode surface (pristine or contaminated with passivation products). Indeed, pristine layers made of reaction with oxygen or water can form a passivation layer, hindering or entirely blocking ionic transport. In addition, contact with the electrolyte may result in the decomposition of the latter into solid products that adhere to the surface of the electrode, in analogy with the formation of the SEI in the field of lithium-based batteries.<sup>[138,139]</sup> While electrolytes developing SEIs are typically compatible with high potential positive electrodes and hence a priori more commercially attractive, reversible plating/stripping will be hindered unless the SEI enables ionic transport or additives can play the role as inhibitors preventing passivation of the surface. A well-accepted approach is using different chloride salts, which play the role of inhibitors for passive film formation. However, such approaches cause severe corrosion problems to other parts of the cell design (current collectors, housing, tabs). Within this scenario, alternative research pathways have been pursued by the research community. For the case of magnesium, the blocking nature

of the SEIs formed was soon identified. Thus efforts focused on using electrolyte solvents with a thermodynamic stability window comprising the redox potential of the metal, amongst which tetrahydrofuran, selected ionic liquids, polymers, and most predominantly glymes.<sup>[140,141]</sup> In contrast, for the case of calcium, with a significantly lower redox potential, reversible plating and stripping have been reported in electrolytes inducing the formation of an SEI,<sup>[142]</sup> with the feasibility of the process being determined by the composition of the SEI, which, in turn, is dependent on the electrolyte salt used.<sup>[143]</sup> SEI layers enabling uniform and high ionic conductivity will favor the formation of a mossy or granular-like multivalent metal (M) deposit by inducing more uniform  $M^{n+}$  saturation levels at the M/SEI interface. Good mechanical properties of the SEI are also desired to accommodate volume changes upon cycling but also to limit dendrite formation due to surface-tension forces.<sup>[144]</sup>

The deposition and stripping process of aluminum in non-aqueous electrolytes exists in chloroaluminate room temperature ionic liquids or a mixture of molten chloride salts (i.e.,  $AlCl_3$ -NaCl-KCl). Chloride ions coordinate aluminum, forming different aluminum-chloride complexes, thus preventing an insulating aluminum oxide passivation film on the surface.<sup>[145]</sup> An electrochemical cell containing aluminum as the negative electrode, a chloride-based electrolyte, and a positive electrode of choice (graphite, organics, or sulfides<sup>[146,147]</sup>) can be cycled up to high current densities without the formation of dendrites.<sup>[148]</sup> Although this is attractive, the use of highly corrosive and water-sensitive  $AlCl_3$  hinders any possible practical application of this cell chemistry and calls for the development of less or completely non-corrosive electrolytes able to dissolve the passivation layer formed on aluminum to enable the plating/stripping process.

The common issue with all multivalent battery concepts is the strong complexation of cations with ligands and solvents, which results in high desolvation energies associated with the cations during the charge transfer reaction. This calls for automated approaches and accelerated procedures to unravel electrolytes having multiple features, that is, enabling highly reversible plating/stripping, involving low desolvation energy, and a suitable electrochemical reaction at the positive electrode (most likely coordination or conversion type). The existence of passivation layers, either native or formed during battery operation, can be determinant for the reversibility of the plating/stripping process and, thus, the feasibility of multivalent batteries. Such layers may also induce additional overpotentials and impact the long-term stability and energy efficiency of such battery concepts.

Altogether, these issues require both careful design of experiments (e.g., using three-electrode cells) to detect the presence and extent of side reactions, if any, coupled to support from computational chemistry to switch from an Edisonian trial and error approach to a rational materials engineering enabling faster discoveries on the field of advanced materials and systems.

Within such a complex scenario, methodological developments in the field of simulation, such as the ML potentials, deep learning approaches, and automated workflows<sup>[149]</sup> described above, are precious tools enabling to both accelerate the discovery of suitable positive electrode materials and new

understanding about the fundamental processes taking place during redox operation, both within the electrolyte and at the interfaces, for example, from the use of integrated computational and experimental workflows and XAI to identify the “deep” descriptors for the interface evolution. The above examples include workflows based on DFT calculations, which autonomously identify candidate electrode materials from inputs involving volume change, thermodynamic stability upon oxidation, OCV at different charge states, and corresponding NEB diffusion energies.<sup>[149]</sup> The computational analysis also helps elucidate the fundamental nature of the multi-electron redox process, either merely cationic/anionic or hybrid.<sup>[129]</sup> Last but not least, AI-assisted modeling aiming at understanding electrolyte speciation and its evolution with concentration and current density are determinants to understand transport limitations in the electrolyte.<sup>[141]</sup> These combined approaches should establish the required fundamental understanding of the cell kinetics, which is crucial since this is one of the main bottlenecks to overcome if multivalent charge carrier batteries with practical power performance are to be developed.

## 9. Metal-Sulfur/Oxygen Batteries

Metal-sulfur batteries have been considered a promising storage system for several decades,<sup>[150]</sup> but materials science challenges have hindered their proliferation. Metal anode and sulfur cathode expose different physical and chemical changes during the continuous discharge/charge process triggering several challenges related to the electrochemical cycling of metal-sulfur batteries. Among them, the most problematic seems to be the stripping and deposition of the metal anode and the reduction and oxidation processes of sulfur. It is well accepted that the reduction of sulfur undergoes some multi-step electrochemical reactions with different polysulfides as intermediates.<sup>[151–153]</sup> Electrolytes enable solubility of active components, including sulfur and polysulfides, whereas polysulfides with long chains typically show the highest solubility in the most commonly used electrolytes.<sup>[154,155]</sup> Two different equilibria are usually determined within the electrochemical cell. The reduction of sulfur can be initiated with the sluggish solubility of sulfur in some solvents. The soluble sulfur mitigates reduction reaction and enables conversion of sulfur to the long-chain polysulfides.<sup>[156]</sup> This reaction step determines the high voltage potential, whose voltage and length depend on the solubility of the long-chain polysulfides. Electrolytes with sparingly soluble polysulfides have a lower potential of high voltage plateau than electrolytes with a high solubility.<sup>[157]</sup> In contrast, high voltage plateaus are absent during sulfur conversion in electrolytes without any sulfur/polysulfide solubility.<sup>[158]</sup> The low voltage plateau is connected with the precipitation of  $Li_2S$  from polysulfides having a mid-chain length. The transition between high and low voltage plateaus represents the shortening of polysulfide chain lengths. Different intermediates have different solubility, electronic conductivity,<sup>[159]</sup> and solubility typically decreases with the shortening of polysulfide chain length. Soluble polysulfides undergo a disproportionation reaction responsible for the polysulfide shuttle mechanism and enable the growth of large particles of the  $Li_2S$  at the

end of discharging or  $S_8$  at the end of charging.<sup>[160]</sup> The electrochemical reduction and oxidation reactions are terminated once the supply of active material (polysulfides trapped outside of the cathode) or electrons (thick deposits of insulating layers) are disabled. In systems where sulfur and polysulfide solubility is entirely disabled, the electrochemical conversion of sulfur to the sulfide/polysulfide mixture proceeds in solid-state conditions. The reaction mechanism shows a single sloping plateau, and the electrochemical reaction is terminated with space restrictions due to volumetric changes caused by the transformation of sulfur into  $Li_2S$ .

These findings yield that the solubility of the active components is crucial for improving the overall performance of metal-sulfur batteries. With different electrolytes at various concentrations of salts and additives leading to different battery performances, a general opinion in the literature persists that the solubility of sulfur and polysulfides is needed to activate solid-state reaction products and adequate kinetics. Solubility can be mitigated with solvents, type and concentration of salts, adsorption additives, catalysts, host matrices, and interlayers. This constitutes a multi-component system that can contribute to high sulfur conversion with fast kinetics and maintain long-term cycling stability in lean electrolyte conditions. The experimental methods related to high-throughput experimentation developed within BATTERY 2030+ aim to rapidly identify experimental evidence for optimal mixtures, compositions, and formulations, and processing.<sup>[28]</sup> Coupled with the autonomous analysis and integrated workflows within BIG-MAP, there will be a major push toward the accelerated discovery of improved materials for metal-sulfur batteries.

Metal-oxygen batteries and (aprotic) lithium-oxygen, in particular, have also been the topic of intense research for more than two decades following the work from Abraham and Jiang in 1996.<sup>[161]</sup> Driven by an alluring theoretical specific energy of  $\approx 3.500 \text{ Wh kg}^{-1}$ , notable progress has been achieved in unraveling the fundamental mechanisms for both surface-based<sup>[162]</sup> and solution-mediated mechanisms.<sup>[163]</sup> However, mass-market commercialization remains elusive due to parasitic processes occurring at the reaction product–electrolyte, product–cathode, electrolyte–cathode, and electrolyte–anode interfaces.<sup>[164,165]</sup>

The formation of highly reactive superoxide,  $Li_2O_{2-x}$  species, and singlet oxygen in particular<sup>[166,167]</sup> poses a key challenge for the realization of commercially viable  $Li-O_2$  batteries and accelerated procedures for identifying mitigating strategies like suitable singlet oxygen quenchers or oxygen blocking SEI<sup>[168]</sup> are needed. The methodological toolkit developed in BATTERY 2030+ and related initiatives offer an opportunity to accelerate the identification and implementation of approaches like BIG-BMS,<sup>[4]</sup> where multisensory data (gas, pressure, etc.) from the embedded sensors (e.g., developed in SENSIBAT, INSTABAT, and SPARTACUS) is fed to the deep learned spatio-temporal Battery Interface Genome (BIG) models<sup>[54]</sup> for the preemptive launch of self-healing additives (HIDDEN and BAT4EVER) like singlet oxygen-quenchers. The BIG-BMS models can be trained to predict the onset of such critical events by using autonomous computational workflows developed to predict anionic redox processes, peroxy-species, and formation of molecular oxygen in Li-rich compounds.<sup>[169]</sup>

Lowering the charging potential through the use of redox mediators (RM) has also shown promise toward limiting the degradation processes,<sup>[170,171]</sup> but the underlying physical principles still need to be fully understood.<sup>[165]</sup> The search for more efficient RMs<sup>[172]</sup> can be accelerated by molecular embeddings,<sup>[173]</sup> enabling inverse design. Other concepts like aqueous lithium-oxygen<sup>[174]</sup> dual-carbon electrode architectures,<sup>[175]</sup> Na,<sup>[176]</sup> K, Mg, Al, and Zn-oxygen would also benefit from the integrated framework outlined above.

## 10. Transport Processes in Ordered and Disordered Systems

Fast ion transport is a critical condition to achieve relevant rates of ionic insertion in electrode materials and conductivities comparable to liquids in ceramic electrolytes. Identifying the chemical and structural descriptors that govern ion diffusion is the key to effectively screening compositions and structures to find fast ion conductors.

In intercalation materials, mobile ions are reversibly incorporated into vacant lattice sites without major atomic rearrangement, either through the nucleation of a second phase with different composition (two-phase reactions) or through the formation of a non-stoichiometric compound whose composition evolves throughout the process (solid solution). Open structures capable of accommodating different ion sizes with minor volume changes can be suitable host materials, although other factors such as ligand interaction and the diffusion topology are key.<sup>[177]</sup>

Migration energies are controlled by the relative site energies (initial, intermediate, final) along the diffusion path as ions hop from one site to another, typically through face-shared intermediate sites.<sup>[178–181]</sup> Migration will be favored if both end configurations and intermediate sites match the cation preferred coordination (or at least with minimal coordination change along the diffusion path) or penalized if significantly different.<sup>[182]</sup> It has been established that, despite  $Na^+$  ions being larger than  $Li^+$ , they can diffuse faster in certain layered structures as their ability to accommodate larger cations as  $Na^+$  results in a larger interlayer spacing that reduces the alkali migration barrier.<sup>[183]</sup> On the other hand, multivalent compounds generally exhibit lower mobility because the higher charge polarizes the host environment. However, reasonable migration barriers can still be obtained by pairing the diffusion topology of a structure with the site preference of each intercalant.<sup>[182]</sup> Together with thermodynamic properties (capacity and voltage), these requirements lead to very specific design criteria for cathode materials with high-rate capability. This is particularly arduous for multivalent cathodes, where one of the main challenges resides in identifying the proper host/intercalant pairs. Automated workflows incorporating physical-based insight using high-throughput semi-empirical (force fields) and ab initio (density functional theory) calculations will define the materials space to be explored and spot promising compounds and compositions, yield the targeted properties based on intertwined structural and compositional descriptors.

Order (i.e., the absence of disorder) has traditionally been a paradigm in intercalation chemistry, as larger activation



barriers<sup>[184]</sup> or lower diffusivities<sup>[185]</sup> have been obtained in materials with antisite defects (i.e., when the TM and the mobile ions are intermixed). However, if fully understood and controlled, the disorder can provide a depth of control and utility to design better materials. For example, identifying when a percolation network providing specific lithium transport pathways is formed in disordered rock-salt cathodes once enough Li excess is introduced.<sup>[186]</sup> Such findings broaden the chemical space to design more performant materials, and ML-assisted phase space exploration and property conditioned inverse generative models represent powerful tools to guide and accelerate new developments.

Unlike unsubstituted and fully ordered systems, substituted or disordered systems need larger simulation boxes and require massive statistical sampling of atomic configurations for each composition and degree of intercalation (for electrode materials). Beyond energies, properties like diffusion path kinetics have a much higher computational cost. Even with state-of-the-art supercomputing and workflow infrastructure, it is not practically feasible to systematically explore target phase space for disordered systems with DFT. Machine learning techniques using local descriptors for on-the-fly assessment of diffusion barriers<sup>[17]</sup> as a function of lithiation levels in disordered systems based on structures obtained from cluster expansion<sup>[187]</sup> or neural-network and other ML-based potentials<sup>[18]</sup> can enable longer time and length scale simulations to capture the structural evolution and diffusion in disordered materials and identify kinetic barrier descriptors/fingerprints. Using, for example, kinetic Monte Carlo methods working with such potentials, it is possible to estimate kinetic properties like power densities in disordered battery electrodes with sufficient accuracy to predict trends and optimize materials composition and utilization. Machine learning models can provide access to diffusion percolation networks<sup>[188]</sup> in disordered materials utilizing the predicted electron density of millions of possible disordered structures.<sup>[36]</sup> Even with fast ML potentials, it is not possible to thoroughly explore the disordered material phase space. Thus, using generative models like VAE built with DFT and ML-potential derived data to inverse design new materials simultaneously will enable co-optimization of all the target properties and facilitate the creation of designer disordered materials.

The structural considerations for ionic diffusion discussed above apply as well for ceramic electrolytes. The difference with respect to intercalation materials is that the concentration of mobile species is kept constant. Still, transport requirements are far more stringent than for electrode materials, which require large carrier densities. Therefore, typical superionic conductors exhibit a vast number of empty or partially filled crystallographic sites. Aliovalent substitutions represent a common strategy to find the best balance between high carrier concentrations and a disordered cation substructure. Other strategies include changing the nature of the anion or the poly-anionic group. Moreover, substitutions can also modify the size of the bottleneck pathways or sintering temperatures and calcination times.<sup>[189]</sup> Exploring the chemical space to engineer the static lattice and elucidate its impact on the ion migration, stability, and processability of ionic conductors represents a very

active field of research that requires a tremendous effort<sup>[190]</sup> and can be greatly assisted by automated and autonomous decision-making experimental material-development platforms.

## 11. Transport across Solid/Solid and Solid/Liquid Interfaces

Fast ionic transport across stable battery interfaces is essential in essentially all types of battery systems. Different types of interfaces can be found depending on the battery technology (liquid/solid, solid/solid, gas/solid) that results in different activation barriers and interfacial resistances.<sup>[191]</sup> The most evident are those between the electrodes and the electrolyte, but there are also “buried” interfaces between active materials and additives or current collectors within composite electrodes.

Today, most commercial battery technologies involve using solid electrodes and liquid electrolytes (either aqueous or organic). Fast interface kinetics is still expected even when organic-solvent-based electrolytes are considered, with an ionic conductivity much lower than the aqueous counterparts. The main limitation can come from the desolvation of ions at the interface, as the complexes formed with the electrolyte solvent molecules may involve strong interactions, especially in multi-valent species. The activation barriers for the interfacial charge transfer are also modulated by the Lewis basicity of the solvent, together with salt concentration (and hence ion pairing).

One specific case of such batteries is those in which the electrode's operation potential lies outside the electrolyte's thermodynamic stability window. A passivation SEI layer is formed at the interface, through which ionic transport can be hindered. The SEI takes a dual functionality: to block further electron-electrolyte interaction to prevent additional electrolyte decomposition and allow for ionic transport (which encompasses ion de-solvating at the SEI/electrolyte interface, diffusion through the SEI layer, and combination with the electrons at the electrode/SEI interface). The design of suitable SEI layers through the formulation of multi-solvent and multi-salt electrolytes still represents a major challenge for most emerging battery chemistries, including the practical realization of silicon anode-based LIBs.<sup>[192]</sup> Major unknowns comprise the understanding of structure-property correlations and the layer formation, growth, and evolution mechanisms.<sup>[193]</sup> Despite progress over the years, the complex nature of the SEI (thinness, multi-layered and multi-component structure) remains to be fully understood, and the lack of tools to directly probe its physical properties and the role of contaminants (e.g., H<sub>2</sub>O traces and decomposition products that can also trigger cascade reactions) make predictive modeling extremely complex. AI-assisted interpretation of experimental results coupled with methods like the hierarchical multi-scale VAEs discussed above should allow outpacing the established understanding of the SEI. With the recognized mechanisms and deep descriptors (reduction voltage, Li<sup>+</sup> binding energies, etc.), SEI formation will be able to shift from a classical trial-and-error process to directed “in vitro” (i.e., artificial coating before cell assembly) or “in vivo” (i.e., in situ modification) SEI design.

Other types of solid/solid interfaces comprise those between electrode materials and solid electrolytes (SE). Interfacial

problems account for much of the hardship faced in the course of realizing all-solid-state batteries. Besides poor contact between the electrodes and the SE (pores, voids formed during cell operation), other significant sources of charge accumulation and mass transfer resistance arise from grain boundaries, chemical and electrochemical reactions at the interfaces. Electrode materials can operate outside the stability window of SE, resulting in decomposition products and interfacial reactions with active materials whose nature will depend on the specific combination of materials (including inactive materials such as carbon) and the electrode state of charge. The use of thin protective coating layers acting as artificial SEI (or similarly the CEI at the cathode) is a common strategy to minimize chemical and electrochemical reactions at solid/solid interfaces. Compositional screening of electrode materials-(coatings)-SE combinations thus become a priority for a suitable selection of materials.<sup>[194]</sup> However, the kinetics of the reactivity at the interface (activation barriers for bond making/breaking and interface diffusion) should be considered in addition to the thermodynamics. The ML interface potentials described above will provide predictive accuracy across multi time- and length-scales, and enable control of the decomposition kinetics of chemical and electrochemical reactions so that stable conducting interfaces that will allow practical all-solid-state batteries can be built.

## 12. Organic Battery Materials

Redox-active organic materials represent an interesting group of future sustainable, low-cost materials with the potential to be used in different applications, mainly where the high volumetric energy density is not a prerequisite.<sup>[195–198]</sup> They consist of abundant elements (C, H, N, O, and S) and can be prepared from, for example, biomass using synthesis techniques with a low CO<sub>2</sub> footprint. Different types of redox-active organic materials have been proposed for the application in batteries, ranging from dissolved small molecules in redox flow batteries and all-solid-state batteries to organic salts and polymers, which can be cycled in the batteries containing liquid electrolytes.<sup>[199–202]</sup>

Essentially all the discussed battery systems can be assembled in the all-organic battery configuration or the combination with different metal electrodes. The versatility of organic chemistry and the need for higher capacity (low molecular weight) opens a large podium of different configurations and possibilities, and their performance should be carefully checked. That is recently more actively performed with computational modeling, which can predict properties of novel redox couples with enhanced performance or stability,<sup>[203]</sup> and with the development of machine learning<sup>[204]</sup> and advanced robotics.<sup>[205]</sup>

There are two types of redox-active organic materials, n-type where cations coordinate the redox centers, and p-type, where the redox centers are coordinated by anions.<sup>[206]</sup> In rare cases, the material can exhibit bipolar functionality, meaning that both anions and cations coordinate redox centers. Weakly bonded molecules or molecular chains enable easy and fast diffusion of electroactive species used for charge compensation, including bulky anions and partially desolvated cations.<sup>[207,208]</sup>

This implies the need to study electrolyte properties along with the development of organic redox materials. Other important parameters for batteries like the specific capacity, working voltage, rate capability, and cycling stability depend on the physicochemical properties of the redox-active moieties. For instance, the ratio between molecular weight and number of redox centers, the position of redox centers in the molecule, electron-withdrawing and electron acceptor groups, polymerization, and wiring (electronic and ionic conductivity) influence the electrochemical properties of materials and consequently their suitability in battery systems.

This calls for a harmonized approach based on computational techniques and designed experimental work to enable faster development and precise identification of a promising combination of redox-active materials and electrolytes.<sup>[209]</sup>

Key points for improvement are enhanced performance and extended durability. Computational approaches to accelerate the design and discovery of more durable organic battery materials like symmetric redox couples for organic flow batteries<sup>[203]</sup> is one of the areas where the development of ML-based methods has been most prolific using, for example, VAEs,<sup>[45]</sup> GANs,<sup>[210]</sup> recurrent neural networks,<sup>[211]</sup> deep RL<sup>[48]</sup> and genetic algorithms (GAs).<sup>[212]</sup> New methods for the accelerated exploration of chemical compound space using, for example, ensemble representation methods like Free energy Machine Learning (FML)<sup>[213]</sup> and curiosity-based approaches not only enable fast prediction of molecular properties with chemical accuracy but also transition states<sup>[214]</sup> and (electro)chemical reaction rates.<sup>[215]</sup> This is a critical step forward in the design of complex organic battery systems such as full electrolyte formulations and the description of degradation reactions of redox molecules in solution, and the development of workflows for training neural network potentials for organic electrolytes and electrode-electrolyte interfaces is central in BIG-MAP and other BATTERY 2030+ projects.

## 13. Summary and Outlook

BATTERY 2030+ has embarked on a mission to develop a versatile and chemistry-neutral platform for the accelerated discovery of novel battery materials and interfaces. While the initial focus in the six research projects is on demonstrating the capabilities of next-generation Li-ion batteries, the machine and deep learning methodologies, autonomous orchestration software, and integrated research infrastructure is developed as modular and externalizable tools, which will enable accelerated discovery and development of emerging, low-TRL ultra-high performant battery concepts and designs. Here, we have illustrated how the AI-based modules and tools being developed to address the main challenges for “nearest in time chemistries” (European SET-Plan action 7<sup>[216]</sup>), (e.g., generalizable workflows and recipes for developing NNPs for long time- and length-scale predictions of the evolution of the battery SEI), can also be applied to resolve the most pertinent challenges for emerging low-TRL battery concepts and designs. Using a representative set of examples, that is, multivalent anodes, metal-sulfur/oxygen batteries, ordered and disordered systems, bulk vs. interface-limited batteries, and organic battery materials, we

identify their main challenges and outline how these can be addressed with the AI-based toolkits and platform(s) currently under development.

The methodological developments within explainable AI and the closed-loop infrastructure will play a critical role in accelerating the discovery process for emerging battery technologies and concepts. It will enable accelerated identification of the limiting processes and deep descriptors. It is important to stress that the critical transition from today's partially automated to the fully autonomous discovery process will depend heavily on the modularity and externalizability of the developed AI-assisted tools. It should also be noted that the success will depend critically on the methods themselves, as well as the availability of (very) large curated and FAIR datasets from all parts of the discovery cycle. The latter is critical to get sufficient data to obtain reliable statistics to derive hyperparameters and descriptors of the materials in their more complicated electrochemical environments. Developing physics- and uncertainty-aware data-driven methods<sup>[217]</sup> capable of training on such multi-sourced experimental and simulation data will strongly enhance the quality of the deep interface descriptors and features that play a critical role in shortening the path to realizing emerging battery technologies and concepts.

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## Conflict of Interest

The authors declare no conflict of interest.

## Keywords

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