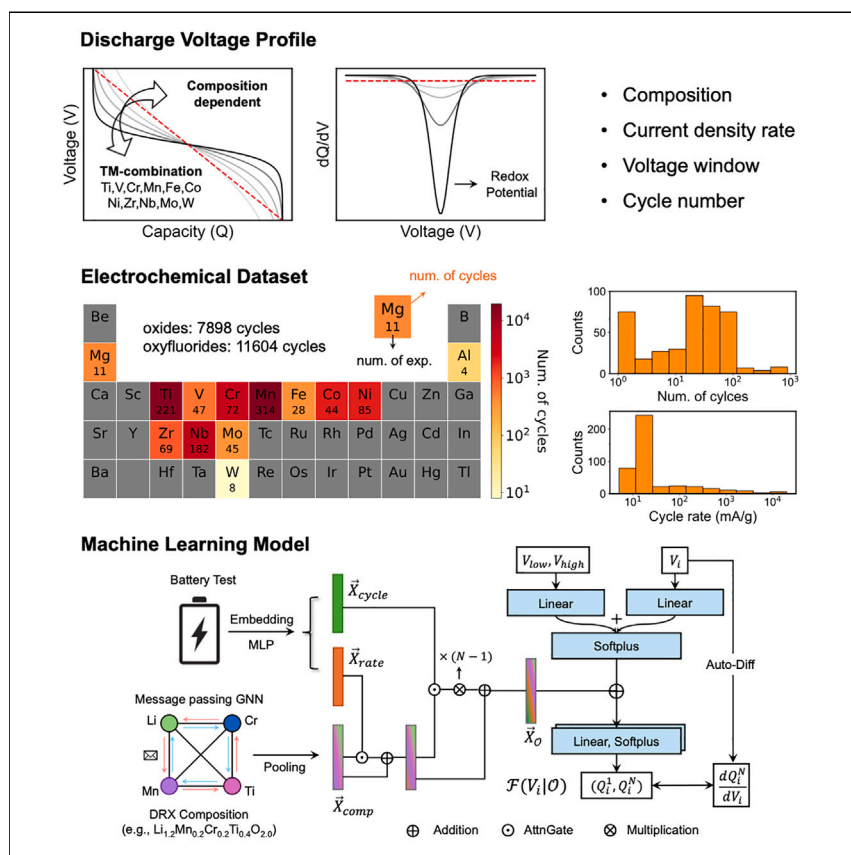


Article

# Deep learning of experimental electrochemistry for battery cathodes across diverse compositions



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Highlights

Machine learning battery informatics across diverse compositions

Representation learning of cathode electrochemistry from experiments

An open-source dataset of discharge voltage profiles

AI has shown promise in battery informatics, but its application to cathodes is limited by complexity and data scarcity. A machine learning model, DRXNet, is developed by training on an extensive experimental electrochemistry dataset encompassing diverse compositions of disordered rocksalt cathodes. DRXNet can predict discharge voltage profiles based on a given composition and test conditions. This data-driven approach provides a heuristic solution to the rapid identification of novel cathode materials, accelerating the development of next-generation batteries.

Article

# Deep learning of experimental electrochemistry for battery cathodes across diverse compositions

Peichen Zhong,<sup>1,2,\*</sup> Bowen Deng,<sup>1,2</sup> Tanjin He,<sup>1,2</sup> Zhengyan Lun,<sup>1,3</sup> and Gerbrand Ceder<sup>1,2,4,\*</sup>

## SUMMARY

Artificial intelligence (AI) has emerged as a tool for discovering and optimizing novel battery materials. However, the adoption of AI in battery cathode representation and discovery is still limited due to the complexity of optimizing multiple performance properties and the scarcity of high-fidelity data. We present a machine learning model (DRXNet) for battery informatics and demonstrate the application in the discovery and optimization of disordered rocksalt (DRX) cathode materials. We have compiled the electrochemistry data of DRX cathodes over the past 5 years, resulting in a dataset of more than 19,000 discharge voltage profiles on diverse chemistries spanning 14 different metal species. Learning from this extensive dataset, our DRXNet model can capture critical features in the cycling curves of DRX cathodes under various conditions. Our approach offers a data-driven solution to facilitate the rapid identification of novel cathode materials, accelerating the development of next-generation batteries for carbon neutralization.

## INTRODUCTION

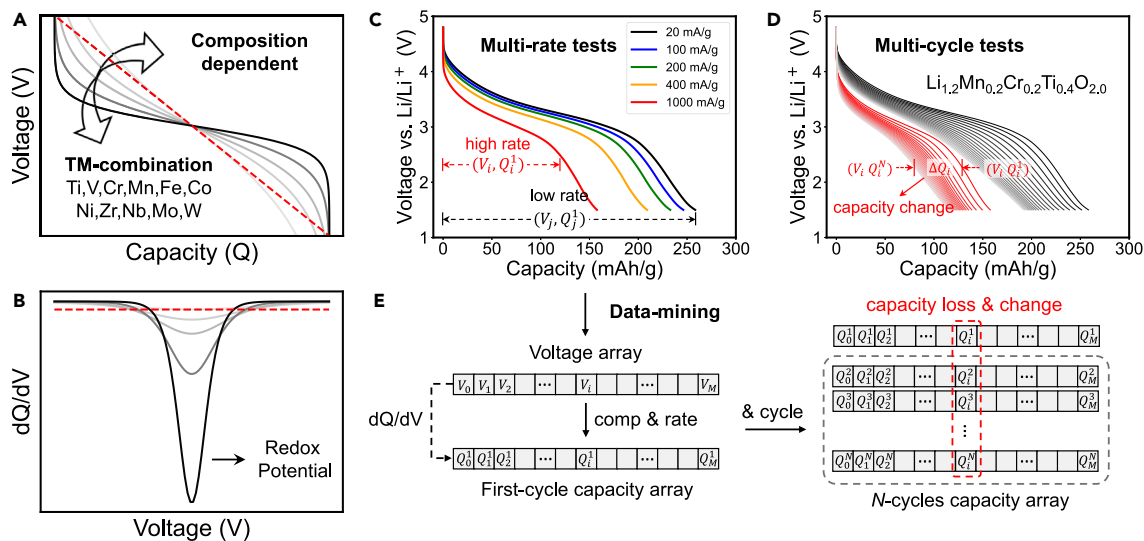
The pursuit of carbon neutrality has become a global imperative in the face of climate change, driving the transition to renewable energy sources and the widespread adoption of electric vehicles.<sup>1–3</sup> High-performance battery cathode materials with large energy density, high-rate performance, and long cycle life are central to these advancements. The development of new cathode materials is essential for meeting the increasing demand for energy storage and advancing the electrification of transportation systems.<sup>4</sup>

Artificial intelligence (AI) has emerged as a potential tool in the discovery and optimization of novel battery materials.<sup>5,6</sup> By leveraging vast amounts of experimental and computational data, AI-assisted techniques can accelerate the design and synthesis of battery materials by identifying promising candidates within large chemical spaces,<sup>7–9</sup> uncovering hidden structure-property relationships via machine-learned atomistic modeling,<sup>10</sup> predicting the remaining lifespan of batteries,<sup>11–15</sup> and optimizing protocols for fast charge/discharge protocol.<sup>16</sup> These efforts significantly reduce the time and cost required for conventional trial-and-error approaches. Most recently, a battery data genome initiative has been proposed to use AI assistance to accelerate the discovery and optimization of battery materials.<sup>17</sup>

Despite these advancements, current machine learning efforts in battery research primarily focus on predicting the lifespan for a simple chemistry or within a limited chemical space, such as Ni-Mn-Co (NMC) or LiFePO<sub>4</sub> (LFP). The development of exploratory machine learning for representing comprehensive compositional effects

## CONTEXT & SCALE

Artificial intelligence (AI) has emerged as a tool for discovering and optimizing novel battery materials. Most machine learning approaches predicting battery performance have been focused on predictions for a specific chemistry or limited chemical space of commercialized cathodes due to the complexity of optimizing multiple performance properties simultaneously. Recent studies in computational material science have demonstrated the feasibility of building universal models for atomistic modeling by harnessing more than 10 years of *ab initio* calculations spanning the periodic table. It becomes a logical extension to envision universal models for the experimental discovery of battery materials. We present a machine learning model that uses an end-to-end training pipeline to encode and learn the (electro)chemical information from experimental voltage profiles. Our approach offers a data-driven solution to facilitate the rapid identification of novel cathode materials.



**Figure 1. Discharge voltage profiles from experiments**

(A) The discharge voltage profile illustrates the relationship between capacity  $Q$  and voltage  $V$ , which is conditioned on the composition of the cathode material.

(B) The derivative quantity  $dQ/dV$  is used to quantify the redox potentials of the TM.

(C and D) The experimental discharge voltage profiles of  $\text{Li}_{1.2}\text{Mn}_{0.2}\text{Cr}_{0.2}\text{Ti}_{0.4}\text{O}_{2.0}$  DRX with (C) multi-rate tests from 20 to 1,000 mA/g and (D) multi-cycle tests from the 1st to the 30th cycle.

(E) The parsed discharge profile is stored in a voltage array  $\{V_i^N\}$  and a capacity array  $\{Q_i^N\}$ , where the subscript  $i$  represents a point (state) on a discharge profile, and the superscript  $N$  represents the cycle number.

in a multi-dimensional chemical space remains underdeveloped due to the challenges associated with simultaneously optimizing multiple electrochemical properties (e.g., rate capability, cyclability, and various test voltage windows).<sup>18</sup> Moreover, the scarcity of high-fidelity data further hinders the progress of AI in the battery field.

Disordered rocksalt (DRX) materials have emerged as promising cathode materials that make use of earth-abundant precursors to enable scaling of Li-ion energy storage to several TWh/year production.<sup>19</sup> Owing to the nearly unlimited compositional design space and considerably more complex structure-property relationship of DRX cathodes, compared with conventional layered cathodes (Figure 1A), their rational design requires the extensive involvement of advanced characterization techniques (e.g., pair-distribution function analysis,<sup>20</sup> spherical-aberration-corrected transmission electron microscopy,<sup>21</sup> and solid-state nuclear magnetic resonance spectroscopy<sup>22</sup>) as well as sophisticated computational tools (e.g., high-dimensional cluster expansion and Monte Carlo simulation<sup>23,24</sup>). Data-driven methods offer alternative means of compositional design and optimization of materials without having to fully construct their structure-property relationships.

In light of these challenges, we developed DRXNet, an exploratory machine learning model for the discovery and optimization of battery cathode materials. DRXNet uses composition, test current density, working voltage window, and cycle number as inputs to predict entire discharge voltage profiles. By training and testing over 19,000 experimental discharge voltage profiles of DRX materials comprising various metal species, we show that the model accurately captures the cathode electrochemistry under different test conditions. Notably, DRXNet captures accessible discharge capacity in diverse Li-Mn-O-F compositions and makes rational predictions for several high-entropy (HE) systems. As a universal model trained on diverse chemistries,

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DRXNet offers a data-driven solution to facilitate the rapid identification of novel cathode materials with improved energy-storage capabilities.

## RESULTS

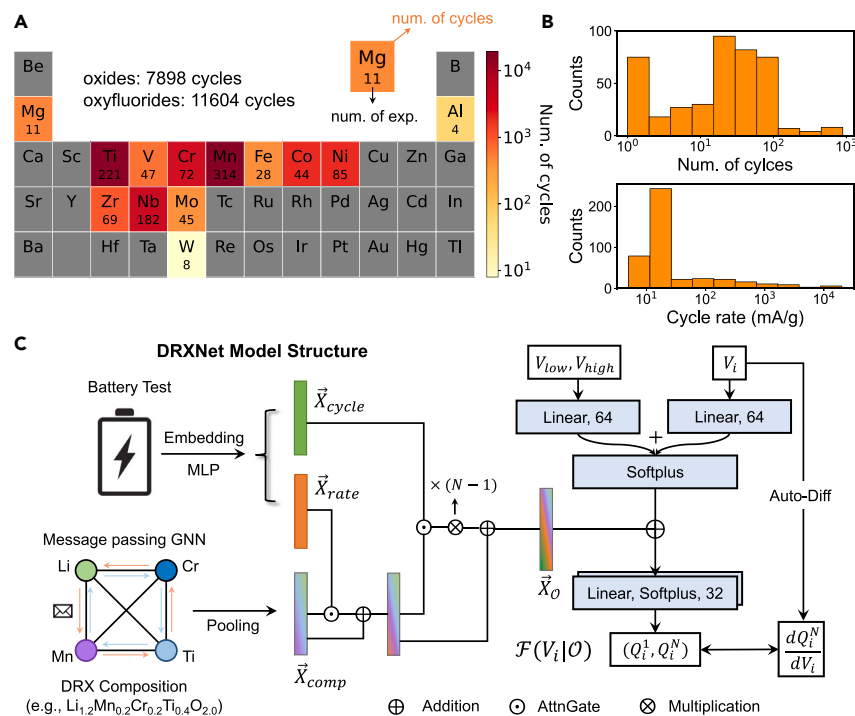
### Parsing discharge profiles

Unlike conventional NMC-based layered cathodes,<sup>25,26</sup> DRX materials exhibit more diverse electrochemical behavior due to the significantly larger chemical space over which they can exist and their more subtle structure involving various forms of cation short-range order (SRO).<sup>27</sup> A prototype DRX cathode ( $\text{Li}_{1+x} \text{M}'_a \text{M}''_b \text{O}_{2-y} \text{F}_y$ ) is composed of three primary compositional parameters: (1) the redox-active species  $\text{M}'$ ; (2) the inert high-valent transition metal  $\text{M}''$ , which charge-compensates for the lithium (Li) excess and stabilizes disordered structures<sup>28</sup>; and (3) fluorine, which enhances the cyclability and accommodates more Li excess without losing transition metal (TM) redox by reducing the anion valence.<sup>29</sup> In addition, other compositional modifications are often made to enhance capacity, rate, or cyclability. For instance, Mg doping in Mn-based oxyfluoride DRX can increase the discharge capacity while retaining a similar voltage-profile shape<sup>30</sup>; Cr doping in  $\text{Li}_{1.2} \text{Mn}_{0.4} \text{Ti}_{0.4} \text{O}_{0.2}$  results in a comparable low-rate capacity but significantly improves the high-rate performance near the top of charge.<sup>31</sup> These non-linear effects arising from compositional changes make both material design and machine learning modeling challenging, thereby necessitating a comprehensive, high-fidelity dataset to address such issues.

Figure 1A introduces the typical discharge voltage profile in battery tests. The profile shape is tied to various factors, such as the DRX composition, applied current density rate, and degradation that may have occurred in prior cycles. Figures 1C and 1D show the multi-rate tests (the first cycle) and multi-cycle tests (of 20 and 1,000 mA/g) of the  $\text{Li}_{1.2} \text{Mn}_{0.2} \text{Cr}_{0.2} \text{Ti}_{0.4} \text{O}_{0.2}$  cathode as an example. The capacity  $Q$  is measured in experiments by determining the cumulative charge transferred in a galvanostatic test. Taking the derivative of  $Q$  with respect to  $V$ , the  $dQ/dV$  value can be evaluated for a given voltage profile, which is a crucial physical quantity for analyzing characteristic redox potentials from different TMs.<sup>32</sup>

### DRX battery TD

We have compiled the electrochemical test data related to DRX compounds by mining electronic experimental notebooks in our research group over the past 5 years to construct the DRX test dataset (DRX-TD). The dataset contains not only results on successful materials published in several papers<sup>27,30,31,33–38</sup> but also data on less well-performing DRX compounds. This endeavor yielded a comprehensive dataset containing 19,000 discharge profiles across 16 different elements (14 metal species + O and F) from lab experiments and published literature (see [experimental procedures](#)). An individual electrochemical test is defined as a group of  $N_{\text{cycle}}$  discharge profiles with a fixed current density rate, where  $N_{\text{cycle}}$  is the number of cycles conducted in such a test, corresponding to the results obtained from one coin cell. The distribution of elements in the DRX-TD is shown in Figure 2A, where the number in each element's box represents the number of compounds with that element present for which an electrochemical test is present. The box's color indicates the total number of discharge profiles for compounds containing that element. Comprising 7,898 discharge profiles of DRX oxides and 11,604 discharge profiles of oxyfluorides, the dataset offers extensive coverage of major redox-active TMs. Figure 2B displays histograms for the number of cycles,  $N_{\text{cycle}}$ , and the current rates at which experiments were performed. As is typical for exploratory research programs in a research laboratory, most of the electrochemical tests were conducted at a low current rate (20 mA/g) and for 10–100 cycles.



**Figure 2. Description of the collected experimental dataset and model design**

(A) The elemental distribution of collected experimental electrochemistry data. The dataset contains 7,898 discharge profiles collected from DRX oxides and 11,604 discharge profiles from oxyfluorides. The color-coding of the boxes indicates the number of discharge profiles (cycles) on compounds that contain that specific element. The number within each elemental box represents the number of compounds with that element on which experiments were conducted.

(B) A histogram of the number of cycles ( $N_{cycle}$ ) and current density (rate) for all the individual electrochemical tests.

(C) An end-to-end pipeline that maps  $Q_i = \mathcal{F}(V_i|\mathcal{O})$ , which consists of the electrochemical condition network  $\mathcal{O}$  (left) and the state prediction network  $\mathcal{F}$  (right). The electrochemical condition network encodes the DRX composition, current density rate, and cycle information. The three encoded vectors are synthesized through gated MLPs with soft attention to obtain the condition vector  $\vec{X}_{\mathcal{O}}$ .<sup>39</sup> The state prediction is approximated as a forward deep neural network that takes the voltage state  $V_i$  and cycling voltage window  $V_{low}, V_{high}$  as inputs. The encoded condition vector  $\vec{X}_{\mathcal{O}}$  is element-wise added in the hidden layer of  $\mathcal{F}$ . The circled symbols are all element-wise operations. The message-passing graph neural network (GNN) is used for compositional encoding of DRX, adapted from the Roost model.<sup>40</sup>

For each discharge profile, 100 points were uniformly sampled from the values of  $V$  and  $Q$ , resulting in a voltage series  $V = [V_1, V_2, \dots, V_i, \dots]$  and a capacity series  $Q = [Q_1, Q_2, \dots, Q_i, \dots]$ . The  $dQ/dV$  curve was then calculated by differentiating  $Q$  with  $V$ . As  $dQ/dV$  is a more intrinsic property for battery materials, including this value in the modeling allows for a more representative analysis of the electrochemical performance of DRX compounds under various conditions (see [experimental procedures](#)).

### DRXNet architecture

DRXNet aims to draw a connection between chemistry and cathode performance by establishing a mapping between  $V$  and  $Q$  for arbitrary cathode compositions under various test conditions. This idea can be conceptualized as identifying a function  $\mathcal{F}$  that maps cathode parameters and the voltage state  $V_i$  to produce the capacity state  $Q_i$  as an output. The function  $\mathcal{F}$  is conditionally defined by the parameters  $\mathcal{O}$ , which consider the electrode composition, current rate, and cycle number,

**Box 1. Algorithm: the workflow of DRXNet with an example of  $\text{Li}_{1.2}\text{Mn}_{0.2}\text{Cr}_{0.2}\text{Ti}_{0.4}\text{O}_2$**

Condition inputs:

$$\vartheta = \begin{cases} \text{composition} = \text{Li}_{1.2}\text{Mn}_{0.2}\text{Cr}_{0.2}\text{Ti}_{0.4}\text{O}_2 \\ \text{rate} = 20\text{mA/g}, \\ \text{cycle} = 1 \end{cases}$$

Condition outputs:

$$\begin{aligned} \vec{X}_{\vartheta_1} &= \vec{X}_{\text{comp}} + \sigma_{f_1}(\vec{X}_{\text{comp}} \parallel \vec{X}_{\text{rate}}) \cdot f_1(\vec{X}_{\text{comp}} \parallel \vec{X}_{\text{rate}}) \\ \vec{X}_{\vartheta_N} &= \vec{X}_{\vartheta_1} + \sigma_{f_2}(\vec{X}_{\vartheta_1} \parallel \vec{X}_{\text{cycle}}) \cdot f_2(\vec{X}_{\vartheta_1} \parallel \vec{X}_{\text{cycle}}) \cdot \mathbf{W}_n(N-1) \end{aligned}$$

Inputs:  $V = [1.5, \dots, V_i, \dots, 4.8] \rightarrow N$  series

for  $i = 1$  to  $N$  do

$$\text{Compute } Q_i = \mathcal{F}(V_i | \vec{X}_{\vartheta_N})$$

end

Outputs:  $Q = [Q_1, \dots, Q_i, \dots, Q_N]$

$$Q_i = \mathcal{F}(V_i | \vartheta). \quad (\text{Equation 1})$$

We designed DRXNet with two main components, as shown in Figure 2C: (1) an electrochemical condition network that generates a feature vector  $\vec{X}_{\vartheta}$  based on the compound's composition and electrochemical test information and (2) a state prediction network to approximate the discharge state of the cathode as a function of the voltage state,  $Q_i = \mathcal{F}(V_i | \vartheta)$ , given the electrochemical conditional encoding of  $\vartheta$ . For instance, Box 1 demonstrates how DRXNet predicts the first-cycle discharge profile of  $\text{Li}_{1.2}\text{Mn}_{0.2}\text{Cr}_{0.2}\text{Ti}_{0.4}\text{O}_2$  at a current rate of 20 mA/g between 1.5 and 4.8 V.

Initially, three condition inputs (composition, rate, and cycle) are encoded to represent  $\vartheta$ . We use Representation Learning from Stoichiometry (Roost), a graph neural network (GNN) model proposed by Goodall and Lee,<sup>40</sup> for compositional encoding. Roost takes elements as graph nodes and updates the correlation between elements through weighted message passing based on each element's fractional concentration. The nodes are initialized with elemental embedded vectors  $\vec{h}_s$  ( $s$ : species) from mat2vec to capture as much prior chemical information as possible through text mining of previously published literature.<sup>41</sup> Moreover, we consider only the cation species as independent nodes in Roost, treating the anion species information (fluorine) as a mean-field background, i.e.,  $\vec{h}'_{\text{Li}} = \vec{h}_{\text{Li}} + c_{\text{F}} \cdot \vec{h}_{\text{F}}$ , where  $c_{\text{F}}$  is the fractional concentration of fluorine, and  $\vec{h}_{\text{Li/F}}$  is the embedded vector of Li/F. Rate and cycle information is encoded using multi-layer perceptrons (MLPs).

Because the rate and cycle properties are intrinsically affected by the composition, we used gated MLPs with soft attention for electrochemical condition encoding via a hierarchical network structure.<sup>39</sup> The  $\vec{X}_{\vartheta_1} = \vec{X}_{\text{comp}} + \sigma_{f_1}(\vec{X}_{\text{comp}} \parallel \vec{X}_{\text{rate}}) \cdot f_1(\vec{X}_{\text{comp}} \parallel \vec{X}_{\text{rate}})$  is a rate-informed feature vector, where  $\sigma_f$  and  $f$  represent MLPs with different activation functions and  $\parallel$  denotes the concatenation operation. In addition, the cycle-informed vector  $\vec{X}_{\vartheta_N} = \vec{X}_{\vartheta_1} + \sigma_{f_2}(\vec{X}_{\vartheta_1} \parallel \vec{X}_{\text{cycle}}) \cdot f_2(\vec{X}_{\vartheta_1} \parallel \vec{X}_{\text{cycle}}) \cdot \mathbf{W}_n(N-1)$  is linearly dependent on the cycle number with a trainable weight  $\mathbf{W}_n$ . Thus, the feature vector  $\vec{X}_{\vartheta_1}$  is used to represent the 1st cycle, and  $\vec{X}_{\vartheta_N}$  is used to represent the  $N$ -th cycle, respectively.

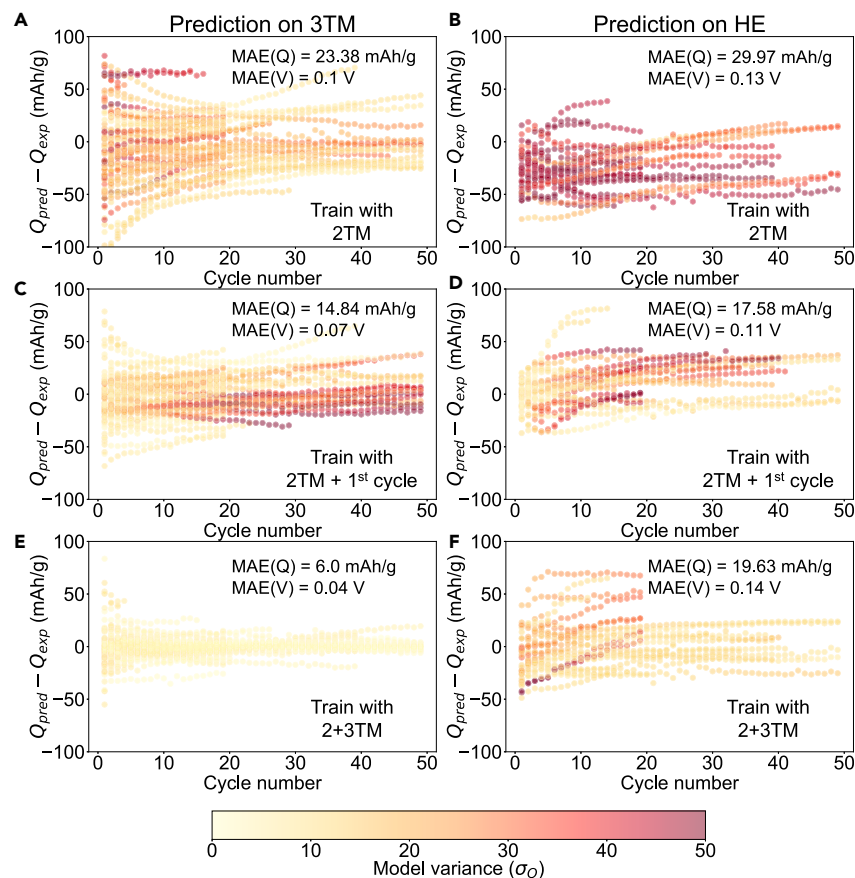
Last, we used several MLPs to construct the state prediction network  $\mathcal{F}$ , as shown in Figure 2C.  $\mathcal{F}$  takes the voltage state  $V_i$  and working window  $V_{\text{low}}, V_{\text{high}}$  as inputs, and the  $\vec{X}_{\mathcal{O}}$  is element-wise added to the hidden layer of  $\mathcal{F}$  to inform  $\mathcal{F}$  of conditions  $\mathcal{O}$  (see experimental procedures). The state prediction network  $\mathcal{F}$  is thus constructed as a simple function mapping from the voltage state  $V_i$  to the capacity  $Q_i$ . In addition,  $(dQ/dV)_i$  is obtained by auto-differentiation of  $\mathcal{F}$ .

### Applicability domain

We explored the scope of DRXNet's applicability in the realm of composition space. Determination of the applicability domain in battery machine learning models can be challenging due to the unavailability of sufficient test data, as generating new data necessitates the synthesis of new materials or conducting battery cycling tests for weeks to months.<sup>18,42</sup> Simply separating the sequence of voltage and capacity signals  $\{V_i, Q_i\}$  into training and test sets can result in data leakage and a failure to represent the expected error in real applications. To evaluate the expressibility and generalization of DRXNet, we designed several experiments by partitioning the dataset based on compositions. The electrochemical tests with no more than two metal species (2TM, excluding Li) were designated as the training set, whereas the tests with three metal species (3TM) and higher numbers of TM components (HE) were assigned as test sets. For each test, an ensemble of five independent models was trained to enhance the overall prediction accuracy and robustness and to quantify the model variance. The average value is used for the prediction, and the standard deviation of the prediction from the ensemble of five DRXNet models ( $\sigma_{\mathcal{O}}$ ) is used to represent the model variance as an approximation of how uncertain the predictions are.

A rational design of battery cathodes typically focuses on the capacity that can be delivered within a certain voltage window. Therefore, we used DRXNet to compute the voltage profiles with electrochemical test parameters in the test set and compared the delivered capacity between 2.0 and 4.4 V of experiments and predictions within 50 cycles (see Figure 3). The voltage range of 2.0–4.4 V (vs. Li<sup>+</sup>/Li) is reasonable for current electrolytes, and most commercialized cathodes such as LFP, LiCoO<sub>2</sub>, and NMC operate within this voltage range. Our choice of this voltage range for testing model performance is aligned with these industry norms. The average voltages ( $\bar{V} = \sum_i V_i \Delta Q_i / \sum_i \Delta Q_i$ ) between 2.0 and 4.4 V were subsequently computed. As a baseline, the mean absolute deviation (MAD) of the average voltage is 0.16/0.21 V for 3TM/HE, and the MAD of discharge capacity is 36.59/38.54 mAh/g for 3TM/HE. Figures 3A and 3B demonstrate the performance of the DRXNet models trained on the 2TM dataset and tested on the 3TM and HE datasets. Mean absolute errors (MAEs) of 0.1/0.13 V for the average voltage and 23.38/29.97 mAh/g for the capacity were obtained for the 3TM/HE TDs, respectively. It is found that large prediction errors already occur for the first cycle and propagate into the subsequent cycles. Notably, a systematic underestimation of capacity is observed for the HE compounds (Figure 3B), which can be rationalized by the fact that 2TM compounds cannot capture the improved performance arising from the novel high-entropy physics.<sup>38,43</sup>

For practical applications, new data points can be continuously collected as experiments progress, enabling on-the-fly training with incoming data to improve predictive performance. To evaluate possible improvement with additional information specific to the system being tested, we evaluated the improvement when DRXNet is trained on a dataset containing all 2TM data and is provided with first-cycle data from 3TM/HE materials. The knowledge of just first-cycle data results in a



**Figure 3. Error and model variance analysis of DRXNet in compositional space**

The prediction error of discharge capacity between 2.0 and 4.4 V (y axis) vs. cycle number (x axis). The model variance is represented by  $\sigma_Q$ , a standard deviation of the ensemble of the models' predictions, which is plotted as scaled colored dots.

(A and B) Predictions on 3TM/HE using models trained on the 2TM dataset.

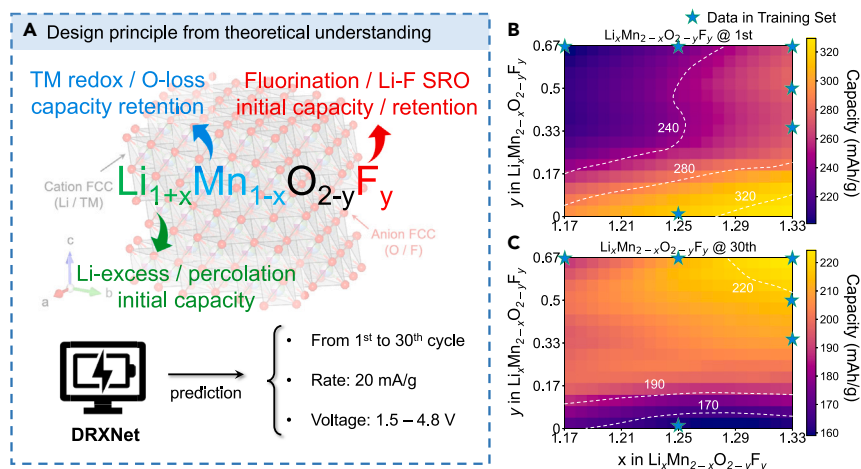
(C and D) Predictions on 3TM/HE using models trained on both the 2TM dataset and the first cycles of the 3TM/HE dataset.

(E and F) Predictions on 3TM/HE using models trained on the 2 + 3TM dataset.

reduction of the mean capacity error from 23.38/29.97 to 14.84/17.58 mAh/g for 3TM/HE (Figures 3C and 3D). The enhanced performance achieved by explicitly training with the first cycle indicates that the model can better generalize cycling performance, even when experiments for a specific composition are not extensively sampled. This capability has the potential to significantly reduce the month-long time frame typically required for electrochemical testing to identify whether a new cathode material has a desired cyclability or rate capability. Training the model with first-cycle data led to a substantial decrease in both the prediction error and model variance for the initial few cycles, although the model variance increased subsequently with the cycle number for untrained domains (Figures 3C and 3D).

To examine how data augmentation could improve the performance of DRXNet, we further trained models on the 2 + 3TM dataset where chemical information, in addition to 2TM interactions, is included. Figures 3E and 3F display the predictions on the 3TM (MAE: 6.0 mAh/g) and HE (MAE: 19.63 mAh/g) datasets. It is important to note that the models trained on 2 + 3TM data show an error reduction of around 10 mAh/g for the HE capacity prediction, compared with the results obtained when





**Figure 4. Illustration of predictions of discharge capacity in Li-Mn-O-F DRX systems**

(A) Compositional design principle includes the optimization of Li-excess content, TM redox, and Li-F short-range order (SRO).<sup>37</sup>

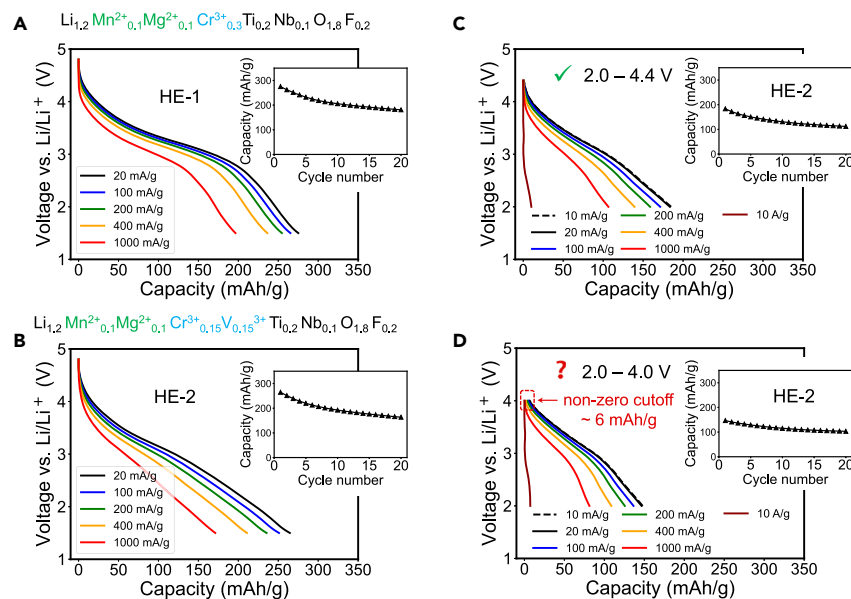
(B and C) Prediction of discharge capacity in the Li-Mn-O-F chemical space for the 1st (B) and 30th (C) cycle between 1.5 and 4.8 V at a current density rate of 20 mA/g. The blue stars indicate the compositions included in the training set.

training the 2TM model (Figures 3B and 3F), along with a significant reduction on the model variance. This finding suggests that the 2TM dataset is inadequate for extracting relevant information and generalizing it to other compositions. The scaling to electrode material with a high number of components necessitates capturing more than 2TM correlations or interactions in training the GNN. Failure to do so may lead to systematic prediction errors, as demonstrated in Figure 3B. When the model is able to acquire sufficient chemical domain knowledge (e.g., 2 + 3TM-model), it becomes feasible to extrapolate the electrochemical properties of high-component electrodes, which is evidenced in Figure 3F with reduced prediction error as well as model variance, and only a few outlier experiments exhibit large errors.

### Recover design principles in Li-Mn-O-F chemical space

We present several examples to illustrate how DRXNet learns the underlying cathode chemistry and assists in designing new materials, where the models used for these applications are pretrained on all discharge profiles. As an attractive earth-abundant, non-precious TM, Mn is of considerable interest for designing next-generation cathode materials.<sup>33</sup> Lun et al. proposed three primary design degrees of freedom for Mn-based DRX (Figure 4A): (1) the Li-excess content, which controls the presence of a percolating network facilitating Li diffusion; (2) the Mn content, as achieving high capacity with a low amount of Mn requires a large amount of oxygen redox, leading to poor cyclability; and (3) the fluorine content, which lowers the total cation valence and provides greater freedom to optimize the Li and Mn content.<sup>37</sup> Fluorine modifies cation SRO through the strong Li-F attraction and lowers the initial capacity,<sup>30,44</sup> but it can increase stability at high voltage charging.<sup>21</sup> These principles are highly correlated and exert non-linear effects on performance.

We used DRXNet to predict the discharge capacity of DRX compounds between 1.5 and 4.8 V at a current rate of 20 mA/g for the 1st and 30th cycles. The results, as a function of Li and F content, are shown in Figures 4B and 4C. The Mn content and valence follow directly from the Li and F content. The effect of fluorine on performance, extensively characterized experimentally, is well captured by our model: a



**Figure 5. Predicted discharge voltage profiles of two high-entropy DRX materials**  
(A)  $\text{Li}_{1.2}\text{Mn}_{0.1}\text{Mg}_{0.1}\text{Cr}_{0.3}\text{Ti}_{0.2}\text{Nb}_{0.1}\text{O}_{1.8}\text{F}_{0.2}$  (HE-1) and (B)  $\text{Li}_{1.2}\text{Mn}_{0.1}\text{Mg}_{0.1}\text{Cr}_{0.15}\text{V}_{0.15}\text{Ti}_{0.2}\text{Nb}_{0.1}\text{O}_{1.8}\text{F}_{0.2}$  (HE-2) with various current densities (from 20 to 1,000 mA/g) between voltage window of 1.5–4.8 V. The inset displays the cycled discharge capacity at a current density of 20 mA/g. HE-2 with various current densities (from 10 mA/g to 10 A/g) between voltage windows of (C) 2.0–4.4 V and (D) 2.0–4.0 V.

higher F content ( $y$  in  $\text{O}_{2-y}\text{F}_y$ ) results in a lower discharge capacity for the 1st cycle but a higher capacity for the 30th cycle, consistent with its documented role in promoting surface stability.<sup>21</sup> In particular, cation-disordered  $\text{Li}_{1.333}\text{Mn}_{0.667}\text{O}_2$  (bottom right corner of Figure 4C) is predicted to have the highest capacity (> 320 mAh/g) for the 1st cycle but the lowest capacity for the 30th cycle. In this compound, the valence of Mn is 4+, and all capacity originates from oxygen. Such a large amount of O-redox leads to rapid capacity fade consistent with the experimental observations on disordered  $\text{Li}_2\text{MnO}_3$  reported in Kataoka et al.<sup>45</sup>

To provide some context for the extrapolation capability of DRXNet, we have illustrated the compositions in the training dataset with blue stars in Figures 4B and 4C. From this, it can be observed that even with a limited distribution of training points on the composition map, DRXNet offers reasonably consistent predictions that seem to be in line with the experimental observations beyond the training points. As DRXNet is trained on various compositions beyond the Li-Mn-O-F chemical space, the ability to extrapolate to other domains can be attributed to the transfer learning from other F- and non-F-containing compounds. The example in this section demonstrates how practitioners can generalize the design principles from a data-driven perspective purely starting from the data mined from experiments.

### Exploratory search for HE cathodes

HE DRXs are composed of many species and present a vast chemical space to explore for battery materials discovery. We used DRXNet for virtual high-throughput screening considering redox-compatible species from the bivalent ( $\text{Mn}^{2+}$ ,  $\text{Fe}^{2+}$ ,  $\text{Ni}^{2+}$ ,  $\text{Mg}^{2+}$ ) and trivalent groups ( $\text{Mn}^{3+}$ ,  $\text{Cr}^{3+}$ ,  $\text{V}^{3+}$ ,  $\text{Fe}^{3+}$ ). Two case studies of predicted HE DRXs are presented:  $\text{Li}_{1.2}\text{Mn}_{0.1}\text{Mg}_{0.1}\text{Cr}_{0.3}\text{Ti}_{0.2}\text{Nb}_{0.1}\text{O}_{1.8}\text{F}_{0.2}$  (HE-1) and  $\text{Li}_{1.2}\text{Mn}_{0.1}\text{Mg}_{0.1}\text{Cr}_{0.15}\text{V}_{0.15}\text{Ti}_{0.2}\text{Nb}_{0.1}\text{O}_{1.8}\text{F}_{0.2}$  (HE-2). The discharge profiles

predicted with DRXNet under various current densities are shown in [Figures 5A](#) and [5B](#). A more comprehensive collection of predictions for other compositions is included in [Figure S6](#).

For HE-1, DRXNet predicts a discharge capacity of 276 mAh/g at a current rate of 20 mA/g. The compound delivers its largest discharge capacity near 3 V and transitions to a higher voltage slope below 3 V, a phenomenon that has been widely observed in Mn redox- and/or Cr redox-based DRXs.<sup>31,38,46</sup> HE-1 is predicted to have an unusually high-rate capability for a DRX compound when discharging. A capacity of 196 mAh/g is estimated at 1,000 mA/g, which is 71% of the capacity at 20 mA/g. Previous work has demonstrated that multi-elemental substitution (i.e., HE strategy) frustrates the unfavorable SRO that leads to poor Li kinetics. In addition, the incorporation of Cr and its migration as Cr<sup>6+</sup> at high voltage create a more extended 0-TM network for Li transport. Both these features improve the Li diffusion kinetics.<sup>31,38</sup> DRXNet clearly learns those benefits and extrapolates rationally into electrochemistry prediction of the HE compositions.

As a comparison to HE-1, we formulated HE-2 with partial V<sup>3+</sup> to Cr<sup>3+</sup> substitution. The change in the shape of the voltage profile, due to the low potential of V<sup>5+</sup>/V<sup>3+</sup> reduction, is well captured by DRXNet as shown in [Figure 5B](#) and  $dQ/dV$  curves in [Figure S7](#). It is clearly demonstrated that with V<sup>3+</sup> incorporation, a nearly constant slope can be observed down to the low-voltage region, which is characteristic for reported V-based DRX cathodes.<sup>47,48</sup> Nevertheless, similar to Cr<sup>6+</sup>, V<sup>5+</sup> can migrate into the tetrahedral sites to enhance Li transport, which benefits the rate capability.<sup>47</sup> Consistently, with this concept, HE-2 is predicted to retain 171 mAh/g capacity at 1,000 mA/g (64% of the 266 mAh/g capacity at 20 mA/g), which is superior to the majority of the DRX cathodes reported to date.

The inset plots in [Figures 5A](#) and [5B](#) show the predicted discharge capacity of HE-1 and HE-2 for 20 cycles. The capacity drop in the first five cycles is predicted to slow down upon further cycling. This result is in full agreement with experimental findings, which indicate some of the irreversibility in the initial cycles, such as cathode-electrolyte interface formation.<sup>49</sup> These examples illustrate how practitioners can effectively use DRXNet to navigate the extensive chemical space of HE DRXs and identify promising candidates for cathode design and optimization.

### Electrochemical conditions

We further tested the depth and transferability of DRXNet's predictive capabilities by varying the HE-2 discharge voltage window and cycling rate, which are typical parameters varied in the investigation of a new cathode material. [Figure 5C](#) displays the discharge profiles between 2.0 and 4.4 V, with two additional rates tested (10 mA/g for a low rate and 10<sup>4</sup> mA/g for an extremely high rate). These conditions are infrequently incorporated into our training data. The 10 mA/g exhibits a discharge profile very similar to that obtained at 20 mA/g, which is entirely consistent with typical experimental findings, as the discharge process at such a low rate exhibits a reduced overpotential and is closer to the equilibrium. The 10 A/g-rate discharge profile demonstrates a sharp drop in voltage, reasonably indicating poor performance at this extremely high rate. Some unphysical predictions start to appear when the model is tested to predict the discharge profiles between 2.0 and 4.0 V. As [Figure 5D](#) shows, a small non-zero offset  $\sim 6$  mAh/g for the 20 mA/g-rate profile appears at the onset of discharge (4.0 V). Since the start of the capacity curve at the upper level of the voltage cutoff is not formally enforced

to zero by the model but emerges from the linear embeddings of the voltage state  $V_i$  with the voltage window  $[V_{\text{low}}, V_{\text{high}}]$ , an offset can be created when there is not enough data for that specific voltage window.

Based on the tests, our primary conclusion is that DRXNet exhibits a reasonable ability to learn the cathode material's chemical information in the latent space and generalize to test conditions that are included in the dataset. However, for test conditions that the model has not or rarely encountered (e.g., experiments with  $V_{\text{high}} < 4.0$  V), discrepancies or unphysical profiles may still arise. This highlights the data scarcity issue, which is typical for human-generated experimental conditions that are biased toward what is needed to demonstrate performance rather than what is optimal for model training.<sup>50</sup>

## DISCUSSION

Most machine learning approaches predicting battery performance have been focused on predictions for a specific chemistry or limited chemical space of commercialized cathodes, typically, the remaining useful life forecasted from the initial cycles.<sup>11,51,52</sup> However, the nature of battery cathode material discovery and optimization lies in a broad domain of chemistries, which is more challenging for AI as it needs to capture the direct (e.g., voltage) and indirect effect (e.g., cycle life) of chemical changes.<sup>18</sup> Recent studies have demonstrated the feasibility of building universal models for atomistic modeling by harnessing more than 10 years of *ab initio* calculations spanning the periodic table.<sup>10,53–56</sup> It becomes a logical extension to envision universal models for the experimental discovery of battery materials by leveraging the wealth of both *ab initio* calculation and experimental data generated on cathode materials worldwide.<sup>17,57</sup> In this work, we propose an end-to-end training pipeline to encode and learn the (electro)chemical information of cathode materials from voltage profiles. Focused on DRX cathodes, we data mined years of lab-generated experimental discharge voltage profiles and trained a universal machine learning model (DRXNet) to make predictions across diverse compositions. This was achieved through a novel model design consisting of an electrochemical condition network  $\mathcal{O}$  and a state prediction network  $\mathcal{F}$ .

The design of the two networks promotes modularity in the architecture, streamlining the optimization and interpretation of each network individually and their learned features. For instance, composition is an intrinsic property of the synthesized cathode materials, and the encoding of such features is independent of other factors such as current density and cycle status, rationalizing our approach to first extract the composition-only feature  $\vec{X}_{\text{comp}}$  via a GNN. Although it remains a challenge that the composition may change as a function of current density and cycle status due to TM dissolution and the irreversible reaction of Li outside the cathode, DRXNet encompasses these factors into the rate- and cycle-informed feature vector representations. By leveraging a ResNet-inspired architecture using skip connections,<sup>58</sup> we achieve a more effective synthesis of the feature vector within the latent space. This design allows for a direct connection between the rate-informed feature,  $\vec{X}_{\mathcal{O},t}$ , and the prediction of the first-cycle capacity. Such architecture has been proven to boost model training and alleviate the well-known gradient vanishing issues.

Given the inherent sequential nature of battery testing data—where possessing information from the  $N$ -th cycle implies the availability of data from the first cycle—it becomes crucial to design features that reflect this causality. This insight leads to the formulation of the cycle-informed feature,  $\vec{X}_{\mathcal{O}_N}$ . This feature accentuates the

difference between the first and  $N$ -th cycles, guiding the prediction for the  $N$ -th-cycle capacity, as detailed in Equation 6. Consequently, our loss function is constructed for multi-task learning with both terms for the first- and  $N$ -th-cycle capacities, ensuring the causal relationships in cycle-dependent capacity predictions (refer to Equation 12). Through an ablation study on whether to include the first-cycle term,  $\ell(Q^1)$ , in the loss function or not, we found that the model without  $\ell(Q^1)$  tends to be underfitted (more details in Figure S8). Our incorporation of loss terms for both the first- and  $N$ -th-cycle capacities enhances the model expressibility, which is a crucial factor in the optimization of battery materials.

In addition, the modular design of the electrochemical condition network ( $\theta$ ) provides flexibility for the feature representation when expanding the model to include other information. The training dataset, derived from our own experimental results, does not encompass testing parameters such as particle size, electrolyte type, synthesis variations, etc. Since the battery electrodes were fabricated in our laboratory using standardized recipes and methodologies, these factors have been coarsely integrated into the compositional model and are treated as constants across our dataset. Currently, the model does not include features to capture structural information (crystal structure, SRO, etc.). In DRX compounds, SRO is known to influence performance, and to the extent that this is not a direct consequence of composition but modified by synthesis parameters, its effects are not accounted for.<sup>59</sup> In principle, researchers can choose to include such factors to design the electrochemical feature vector, depending on the specific problem they are addressing. Given the vast amount and complexity of these properties, a synthetic data collection approach is necessary. Data-mining techniques, such as text mining and figure mining, can automatically retrieve valuable experimental information from decades of published literature,<sup>60,61</sup> although a challenge with aggregating diverse data from literature is the numerous hidden and unspecified variables relevant to materials synthesis and electrochemical testing. Looking forward, autonomous labs can address both data scarcity and transparency issues by enabling more extensive exploration of the experimental space and even better collection of data from “failed” experiments.<sup>62–65</sup>

In conclusion, DRXNet represents a step forward in developing machine learning models for battery materials research. By continuously refining the model and incorporating additional data and parameters, we anticipate that such a machine learning framework will play an increasingly critical role in discovering and optimizing next-generation battery materials.

## EXPERIMENTAL PROCEDURES

### Resource availability

#### Lead contact

Further information and requests for resources and materials should be directed to and will be fulfilled by the lead contact, Gerbrand Ceder ([gceder@berkeley.edu](mailto:gceder@berkeley.edu)).

#### Materials availability

This study did not generate new unique materials.

#### Data and code availability

The codes of DRXNet and the pretrained models are open-sourced at <https://github.com/zhongpc/DRXNet> and <https://doi.org/10.5281/zenodo.10719829>. The open-source dataset is available at <https://doi.org/10.6084/m9.figshare.25328578.v1> for public access, which contains 12,688 experimental discharge voltage profiles excluding the Mn-rich and Ti-based DRX. The open-source dataset is not identical to but rather a part of the DRX-TD that was used for the pretrained models in the paper.

### Data collection

We collected coin-cell electrochemical test data from our lab, starting in 2016, and converted them into a digital format (.json). Each .json file contains information on one individual electrochemical test, including the electrode composition, electrode mass (g), active mass (g), test current rate (mA/g), low and high voltage value of the working window (V), and charge/discharge profiles of  $N_{\text{cycle}}$ -collected cycles. The compositions used for the model training were taken as the targeted composition in experiments. For the fraction of our dataset that was previously published, the composition values were typically confirmed by inductively coupled plasma (ICP) analysis. For these compounds, the feature vectors of the targeted compositions and ICP-analyzed composition exhibit  $\geq 99.7\%$  in cosine similarity as shown in the [supplemental information](#), which supports using the targeted composition for the general prediction purpose. Nonetheless, minor variations between the actual composition and the target composition can be a source of noise in the data.

For the in-house battery tests, the CR2032 coin cells were assembled using commercial 1 M LiPF<sub>6</sub> in an ethylene carbonate and dimethyl carbonate solution (volume ratio 1:1) as the electrolyte, glass microfiber filters (Whatman) as separators, and Li-metal foil (FMC) as the anode. The coin cells were tested on an Arbin battery cycler at room temperature. The cathode consisted of a mixture of active material (DRX), Super C65 carbon black, and polytetrafluoroethylene (PTFE). The capacity signal, collected in units of Ah from the Arbin battery cycler, was normalized to mAh/g using the mass of the active material (active mass). The data from the failed tests (e.g., Arbin cycler breakdown, electrolyte failure, strong signal fluctuations, etc.) were removed from the dataset (see [Figure S1](#) for examples).

To enhance the generalization and expressibility of DRXNet, we expanded the dataset by figure mining published voltage profiles in related systems not covered by our lab tests (see [Table S1](#)), which was accomplished using the WebPlotDigitizer.<sup>66</sup> We used the UnivariateSpline method to denoise and resample the experimental profiles and compute the  $dQ/dV$  curves. One hundred points were uniformly sampled to form the voltage series  $V = [V_0, V_1, \dots, V_i, \dots]$  for each discharge profile, and the capacity series and  $dQ/dV$  series were calculated accordingly from  $V$ .

### Model design

#### Preliminaries

A linear layer with trainable weight  $\mathbf{W}$  and bias  $\mathbf{b}$  is defined as

$$L(\vec{X}) = \vec{X}\mathbf{W} + \mathbf{b}. \quad (\text{Equation 2})$$

For simplicity of notion, each  $L$  represents different trainable weights in the following equations.

#### Compositional encoding

For elemental information, each element is first embedded into a 200-dimensional vector using `mat2vec`.<sup>41</sup> The Roost model is used for compositional encoding,<sup>40</sup> which is a GNN with message passings as follows:

$$\begin{aligned} \vec{h}_i^{t+1} &= \vec{h}_i^t + \sum_{j,m} a_{ij}^{t,m} \cdot \sigma_g \circ L_c \left( \vec{h}_i^t \parallel \vec{h}_j^t \right), \\ a_{ij}^{t,m} &= \frac{w_j \exp \left( e_{ij}^{t,m} \right)}{\sum_k w_k \exp \left( e_{ik}^{t,m} \right)}, e_{ik}^{t,m} = \sigma_g \circ L_a \left( \vec{h}_i^t \parallel \vec{h}_k^t \right). \end{aligned} \quad (\text{Equation 3})$$

In these equations,  $\vec{h}_i^t$  represents the  $t$ -th hidden layer for the  $i$ -th element;  $\parallel$  denotes the concatenation operation; and the soft-attention coefficient  $a_{ij}^{t,m}$  describes the interaction between elements  $i$  and  $j$ , with  $m$  as the index of multi-head attention.  $L_c$  and  $L_a$  denote the linear layer for the core and attention layer, respectively. The fractional concentration  $w_j$  of element  $j$  depends on the specific compound (e.g.,  $w_j = 0.6/0.1/0.1/0.2$  for Li/Mn/Cr/Ti in  $\text{Li}_{1.2}\text{Mn}_{0.2}\text{Cr}_{0.2}\text{Ti}_{0.4}\text{O}_{2.0}$ ).  $\sigma_g$  is the SiLu activation function. After  $n$  graph convolution layers, the encoded composition vector  $\vec{X}_{\text{comp}}$  is obtained by average pooling over the elements with weighted attention:

$$\vec{X}_{\text{comp}} = \text{Pooling} \left[ \frac{w_i \exp \left( \sigma_g \circ L_a \left( \vec{h}_i^n \right) \right)}{\sum_k \exp \left( \sigma_g \circ L_a \left( \vec{h}_k^n \right) \right)} \cdot \left( \sigma_g \circ L_c \left( \vec{h}_i^n \right) \right) \right] \quad (\text{Equation 4})$$

### Electrochemical condition encoding

The electrochemical test primarily involves two pieces of information: the current density rate and cycle number. We use MLPs to encode the rate and cycle number:

$$\vec{X}_{\text{rate}} = \sigma_g \circ L(\text{rate}), \vec{X}_{\text{cycle}} = \sigma_g \circ L(\text{cycle}). \quad (\text{Equation 5})$$

As the actual rate and cycle performance are strongly correlated with cathode materials, the relationship between the composition, rate, and cycle is synthesized using gated MLPs with soft attention<sup>39</sup>:

$$\begin{aligned} \vec{X}_{\phi_1} &= \vec{X}_{\text{comp}} + \sigma_{f_1} \left( \vec{X}_{\text{comp}} \parallel \vec{X}_{\text{rate}} \right) \cdot f_1 \left( \vec{X}_{\text{comp}} \parallel \vec{X}_{\text{rate}} \right) \\ \vec{X}_{\phi_N} &= \vec{X}_{\phi_1} + \sigma_{f_2} \left( \vec{X}_{\phi_1} \parallel \vec{X}_{\text{cycle}} \right) \cdot f_2 \left( \vec{X}_{\phi_1} \parallel \vec{X}_{\text{cycle}} \right) \cdot \mathbf{W}_n (N - 1) \end{aligned} \quad (\text{Equation 6})$$

where  $\sigma_f = \sigma_s \circ B \circ L$  is an MLP,  $\sigma_s$  is the Sigmoid activation function, and  $f = \sigma_g \circ B \circ L$  is an MLP with SiLu activation function  $\sigma_g$ . The BatchNormalization layer  $B$  is added before the activation function. In this equation,  $\vec{X}_{\phi_1}$  is a feature vector jointly determined by the composition and rate information, which is used to predict the first-cycle property.  $\vec{X}_{\phi_N}$  is a feature vector jointly determined by the composition, rate, and cycle information, which is used to predict the  $N$ -th-cycle property. The difference between  $\vec{X}_{\phi_1}$  and  $\vec{X}_{\phi_N}$  is linearly dependent on the number of cycles with a trainable weight  $\mathbf{W}_n$ , allowing the model to learn cycle performance contrastively.

### State prediction network

The state prediction network ( $\mathcal{F}$ ) takes the inputs of voltage state ( $V_i$ ) and outputs the discharge-capacity state ( $Q_i$ ):

$$Q_i = \mathcal{F}(V_i | \theta). \quad (\text{Equation 7})$$

In practice, the voltage profile is measured within the applied voltage window ( $V_{\text{low}}$ ,  $V_{\text{high}}$ ). To accommodate the voltage window in the discharge state prediction, the first layer in  $\mathcal{F}$  is encoded via an MLP:

$$\vec{Z}_i^0 = L \circ \sigma_{\mathcal{F}} \circ [L(V_{\text{low}}, V_{\text{high}}) + L(V_i)], \quad (\text{Equation 8})$$

where  $\sigma_{\mathcal{F}}(\cdot)$  is the Softplus activation function. The test condition information is element-wise added to the state-prediction network.<sup>58</sup>

$$\vec{Z}_i^N = \sigma_{\mathcal{F}} \circ L \left( \vec{Z}_i^0 + \vec{X}_{\phi_N} \right) \quad (\text{Equation 9})$$

The state of capacity is obtained by

$$Q_i^N = \sigma_F \circ L \circ \sigma_F \circ L \left( \vec{Z}_i^N \right) \quad (\text{Equation 10})$$

where  $Q_i^N$  is the capacity for the  $N$ -th cycle (including the first cycle). Because the discharge capacity is always positive,  $\sigma_F$  is added to constrain the predicted capacity to be positive and to accelerate the training process.  $dQ/dV$  for the redox potential can be obtained via PyTorch auto-differentiation<sup>67</sup>:

$$\left. \frac{dQ}{dV} \right|_i = \text{AutoDiff}(Q_i, V_i). \quad (\text{Equation 11})$$

### Model training

The model is trained to minimize the sum of multi-task losses for the capacity of the first cycle, the  $N$ -th cycle, and  $dQ/dV$ :

$$\mathcal{L} = w_Q \ell(Q_i^N) + w_{dQ} \ell\left(\frac{dQ^N}{dV_i}\right) + w_{Q_1} \ell(Q_i^1) + \mathcal{R}. \quad (\text{Equation 12})$$

A MSE loss function is used for  $\ell(Q_i^N)$  and  $\ell\left(\frac{dQ^N}{dV_i}\right)$ , whereas a MAE loss function is employed for the first cycle as a contrastive term  $\ell(Q_i^1)$ . The weights for  $Q_i^N$ ,  $dQ/dV$ , and  $Q_i^1$  are set to  $w_Q = 1$ ,  $w_{dQ} = 1$ , and  $w_{Q_1} = 5$ . The term  $\mathcal{R}$  represents regularization, which consists of two parts: (1) an  $\ell_2$ -norm regularization of the network's parameters  $\|\theta\|_2$  and (2) a smoothing term  $\|dQ/dc\|_2$  to avoid large, unphysical performance fluctuations ( $c$  denotes the fractional concentration of elements). The weight of regularization is  $10^{-4}$ .

To make predictions, an ensemble of five independent models was trained to make predictions. Each model was trained with a batch size of 1,024 within 30 epochs. The Adam optimizer was used with  $10^{-3}$  as the initial learning rate. The ExponentialLR scheduler was used to adjust the learning rate with a decay rate of 0.9 per epoch.

### SUPPLEMENTAL INFORMATION

Supplemental information can be found online at <https://doi.org/10.1016/j.joule.2024.03.010>.

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## AUTHOR CONTRIBUTIONS

P.Z. and G.C. conceived the initial idea. P.Z. collected the dataset and developed the code base with help from B.D. and T.H. Z.L. and G.C. offered insight into the project. P.Z. and G.C. prepared the manuscript. All authors contributed to discussions and approved the manuscript.

## DECLARATION OF INTERESTS

G.C. is a member of the *Joule's* advisory board.

## DECLARATION OF GENERATIVE AI AND AI-ASSISTED TECHNOLOGIES IN THE WRITING PROCESS

During the preparation of this work, the authors used ChatGPT-4 in order to refine the writing. After using this tool, the authors reviewed and edited the content as needed, and they take full responsibility for the content of the publication.

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## REFERENCES

- Olivetti, E.A., Ceder, G., Gaustad, G.G., and Fu, X. (2017). Lithium-Ion Battery Supply Chain Considerations: Analysis of Potential Bottlenecks in Critical Metals. *Joule* 1, 229–243. <https://doi.org/10.1016/j.joule.2017.08.019>.
- Xie, L., Singh, C., Mitter, S.K., Dahleh, M.A., and Oren, S.S. (2021). Toward carbon-neutral electricity and mobility: Is the grid infrastructure ready? *Joule* 5, 1908–1913. <https://doi.org/10.1016/j.joule.2021.06.011>.
- Tian, Y., Zeng, G., Rutt, A., Shi, T., Kim, H., Wang, J., Koettgen, J., Sun, Y., Ouyang, B., Chen, T., et al. (2021). Promises and Challenges of Next-Generation “Beyond Li-ion” Batteries for Electric Vehicles and Grid Decarbonization. *Chem. Rev.* 121, 1623–1669. <https://doi.org/10.1021/acs.chemrev.0c00767>.
- Goodenough, J.B., and Kim, Y. (2010). Challenges for rechargeable Li batteries. *Chem. Mater.* 22, 587–603. <https://doi.org/10.1021/cm901452z>.
- Lv, C., Zhou, X., Zhong, L., Yan, C., Srinivasan, M., Seh, Z.W., Liu, C., Pan, H., Li, S., Wen, Y., et al. (2022). Machine Learning: An Advanced Platform for Materials Development and State Prediction in Lithium-Ion Batteries. *Adv. Mater.* 34, e2101474. <https://doi.org/10.1002/adma.202101474>.
- Wang, H., Fu, T., Du, Y., Gao, W., Huang, K., Liu, Z., Chandak, P., Liu, S., Van Katwyk, P., Deac, A., et al. (2023). Scientific discovery in the age of artificial intelligence. *Nature* 620, 47–60. <https://doi.org/10.1038/s41586-023-06221-2>.
- Ahmad, Z., Xie, T., Maheshwari, C., Grossman, J.C., and Viswanathan, V. (2018). Machine Learning Enabled Computational Screening of Inorganic Solid Electrolytes for Suppression of Dendrite Formation in Lithium Metal Anodes. *ACS Cent. Sci.* 4, 996–1006. <https://doi.org/10.1021/acscentsci.8b00229>.
- Liow, C.H., Kang, H., Kim, S., Na, M., Lee, Y., Baucour, A., Bang, K., Shim, Y., Choe, J., Hwang, G., et al. (2022). Machine learning assisted synthesis of lithium-ion batteries cathode materials. *Nano Energy* 98, 107214. <https://doi.org/10.1016/j.nanoen.2022.107214>.
- Chen, C., Nguyen, D.T., Lee, S.J., Baker, N.A., Karakoti, A.S., Lauw, L., Owen, C., Mueller, K.T., Bilodeau, B.A., Murugesan, V., et al. (2024). Accelerating computational materials discovery with artificial intelligence and cloud high-performance computing: from large-scale screening to experimental validation. Preprint at arXiv. <https://doi.org/10.48550/arXiv.2401.04070>.
- Deng, B., Zhong, P., Jun, K., Riebesell, J., Han, K., Bartel, C.J., and Ceder, G. (2023). CHGNet as a pretrained universal neural network potential for charge-informed atomistic modelling. *Nat. Mach. Intell.* 5, 1031–1041. <https://doi.org/10.1038/s42256-023-00716-3>.
- 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. <https://doi.org/10.1038/s41560-019-0356-8>.
- Jones, P.K., Stimming, U., and Lee, A.A. (2022). Impedance-based forecasting of lithium-ion battery performance amid uneven usage. *Nat. Commun.* 13, 4806. <https://doi.org/10.1038/s41467-022-32422-w>.
- Aykol, M., Gopal, C.B., Anapolsky, A., Herring, P.K., van Vlijmen, B., Berliner, M.D., Bazant, M.Z., Braatz, R.D., Chueh, W.C., and Storey, B.D. (2021). Perspective – Combining Physics and Machine Learning to Predict Battery Lifetime. *J. Electrochem. Soc.* 168, 30525. <https://doi.org/10.1149/1945-7111/abec55>.
- Hu, X., Xu, L., Lin, X., and Pecht, M. (2020). Battery lifetime prognostics. *Joule* 4, 310–346. <https://doi.org/10.1016/j.joule.2019.11.018>.
- Sulzer, V., Mohtat, P., Aitio, A., Lee, S., Yeh, Y.T., Steinbacher, F., Khan, M.U., Lee, J.W., Siegel, J.B., Stefanopoulou, A.G., et al. (2021). The challenge and opportunity of battery lifetime prediction from field data. *Joule* 5, 1934–1955. <https://doi.org/10.1016/j.joule.2021.06.005>.
- 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. <https://doi.org/10.1038/s41586-020-1994-5>.
- Ward, L., Babinec, S., Dufek, E.J., Howey, D.A., Viswanathan, V., Aykol, M., Beck, D.A.C., Blaiszik, B., Chen, B.-R., Crabtree, G., et al. (2022). Principles of the Battery Data Genome. *Joule* 6, 2253–2271. <https://doi.org/10.1016/j.joule.2022.08.008>.
- Sendek, A.D., Ransom, B., Cubuk, E.D., Pellouchoud, L.A., Nanda, J., and Reed, E.J. (2022). Machine Learning Modeling for Accelerated Battery Materials Design in the Small Data Regime. *Adv. Energy Mater.* 12, 2200553. <https://doi.org/10.1002/aenm.202200553>.
- Clément, R.J., Lun, Z., and Ceder, G. (2020). Cation-disordered rocksalt transition metal oxides and oxyfluorides for high energy lithium-ion cathodes. *Energy Environ. Sci.* 13, 345–373. <https://doi.org/10.1039/C9EE02803J>.
- Key, B., Morcrette, M., Tarascon, J.M., and Grey, C.P. (2011). Pair Distribution Function Analysis and Solid State NMR Studies of Silicon Electrodes for Lithium Ion Batteries: Understanding the (De)lithiation Mechanisms.

- J. Am. Chem. Soc. 133, 503–512. <https://doi.org/10.1021/ja108085d>.
21. Li, L., Lun, Z., Chen, D., Yue, Y., Tong, W., Chen, G., Ceder, G., and Wang, C. (2021). Fluorination-Enhanced Surface Stability of Cation-Disordered Rocksalt Cathodes for Li-Ion Batteries. *Adv. Funct. Mater.* 31, 2101888. <https://doi.org/10.1002/adfm.202101888>.
22. Clément, R.J., Kitchaev, D., Lee, J., and Ceder, G. (2018). Short-Range Order and Unusual Modes of Nickel Redox in a Fluorine-Substituted Disordered Rocksalt Oxide Lithium-Ion Cathode. *Chem. Mater.* 30, 6945–6956. <https://doi.org/10.1021/acs.chemmater.8b03794>.
23. Zhong, P., Chen, T., Barroso-Luque, L., Xie, F., and Ceder, G. (2022). An  $l_1/l_2$ -norm regularized regression model for construction of robust cluster expansions in multicomponent systems. *Phys. Rev. B* 106, 24203. <https://doi.org/10.1103/PhysRevB.106.024203>.
24. Barroso-Luque, L., Zhong, P., Yang, J.H., Xie, F., Chen, T., Ouyang, B., and Ceder, G. (2022). Cluster expansions of multicomponent ionic materials: Formalism and methodology. *Phys. Rev. B* 106, 144202. <https://doi.org/10.1103/PhysRevB.106.144202>.
25. Mizushima, K., Jones, P.C., Wiseman, P.J., and Goodenough, J.B. (1980).  $\text{Li}_x\text{CoO}_2$  ( $0 < x < 1$ ): A new cathode material for batteries of high energy density. *Mater. Res. Bull.* 15, 783–789. [https://doi.org/10.1016/0025-5408\(80\)90012-4](https://doi.org/10.1016/0025-5408(80)90012-4).
26. Manthiram, A. (2020). A reflection on lithium-ion battery cathode chemistry. *Nat. Commun.* 11, 1550. <https://doi.org/10.1038/s41467-020-15355-0>.
27. Ji, H., Urban, A., Kitchaev, D.A., Kwon, D.H., Artrith, N., Ophus, C., Huang, W., Cai, Z., Shi, T., Kim, J.C., et al. (2019a). Hidden structural and chemical order controls lithium transport in cation-disordered oxides for rechargeable batteries. *Nat. Commun.* 10, 592. <https://doi.org/10.1038/s41467-019-08490-w>.
28. Urban, A., Abdellahi, A., Dacek, S., Artrith, N., and Ceder, G. (2017). Electronic-Structure Origin of Cation Disorder in Transition-Metal Oxides. *Phys. Rev. Lett.* 119, 176402. <https://doi.org/10.1103/PhysRevLett.119.176402>.
29. Lee, J., Papp, J.K., Clément, R.J., Sallis, S., Kwon, D.H., Shi, T., Yang, W., McCloskey, B.D., and Ceder, G. (2017). Mitigating oxygen loss to improve the cycling performance of high capacity cation-disordered cathode materials. *Nat. Commun.* 8, 981. <https://doi.org/10.1038/s41467-017-01115-0>.
30. Zhong, P., Cai, Z., Zhang, Y., Giovine, R., Ouyang, B., Zeng, G., Chen, Y., Clément, R., Lun, Z., and Ceder, G. (2020). Increasing Capacity in Disordered Rocksalt Cathodes by Mg Doping. *Chem. Mater.* 32, 10728–10736. <https://doi.org/10.1021/acs.chemmater.0c04109>.
31. Huang, J., Zhong, P., Ha, Y., Kwon, D.-h., Crafton, M.J., Tian, Y., Balasubramanian, M., McCloskey, B.D., Yang, W., and Ceder, G. (2021). Non-topotactic reactions enable high rate capability in Li-rich cathode materials. *Nat. Energy* 6, 706–714. <https://doi.org/10.1038/s41560-021-00817-6>.
32. Newman, J., and Balsara, N.P. (2021). *Electrochemical Systems* (John Wiley & Sons).
33. Lee, J., Kitchaev, D.A., Kwon, D.H., Lee, C.W., Papp, J.K., Liu, Y.S., Lun, Z., Clément, R.J., Shi, T., McCloskey, B.D., et al. (2018). Reversible  $\text{Mn}^{2+}/\text{Mn}^{4+}$  double redox in lithium-excess cathode materials. *Nature* 556, 185–190. <https://doi.org/10.1038/s41586-018-0015-4>.
34. Kitchaev, D.A., Lun, Z., Richards, W.D., Ji, H., Clément, R.J., Balasubramanian, M., Kwon, D.-H., Dai, K., Papp, J.K., Lei, T., et al. (2018). Design principles for high transition metal capacity in disordered rocksalt Li-ion cathodes. *Energy Environ. Sci.* 11, 2159–2171. <https://doi.org/10.1039/C8EE00816G>.
35. Lun, Z., Ouyang, B., Kitchaev, D.A., Clément, R.J., Papp, J.K., Balasubramanian, M., Tian, Y., Lei, T., Shi, T., McCloskey, B.D., et al. (2019). Improved Cycling Performance of Li-Excess Cation-Disordered Cathode Materials upon Fluorine Substitution. *Adv. Energy Mater.* 9, 1802959. <https://doi.org/10.1002/aenm.201802959>.
36. Ji, H., Kitchaev, D.A., Lun, Z., Kim, H., Foley, E., Kwon, D.-H., Tian, Y., Balasubramanian, M., Bianchini, M., Cai, Z., et al. (2019b). Computational Investigation and Experimental Realization of Disordered High-Capacity Li-Ion Cathodes Based on Ni Redox. *Chem. Mater.* 31, 2431–2442. <https://doi.org/10.1021/acs.chemmater.8b05096>.
37. Lun, Z., Ouyang, B., Cai, Z., Clément, R.J., Kwon, D.-H., Huang, J., Papp, J.K., Balasubramanian, M., Tian, Y., McCloskey, B.D., et al. (2020). Design Principles for High-Capacity Mn-Based Cation-Disordered Rocksalt Cathodes. *Chem* 6, 153–168. <https://doi.org/10.1016/j.chempr.2019.10.001>.
38. Lun, Z., Ouyang, B., Kwon, D.H., Ha, Y., Foley, E.E., Huang, T.Y., Cai, Z., Kim, H., Balasubramanian, M., Sun, Y., et al. (2021). Cation-disordered rocksalt-type high-entropy cathodes for Li-ion batteries. *Nat. Mater.* 20, 214–221. <https://doi.org/10.1038/s41563-020-00816-0>.
39. Xie, T., and Grossman, J.C. (2018). Crystal Graph Convolutional Neural Networks for an Accurate and Interpretable Prediction of Material Properties. *Phys. Rev. Lett.* 120, 145301. <https://doi.org/10.1103/PhysRevLett.120.145301>.
40. Goodall, R.E.A., and Lee, A.A. (2020). Predicting materials properties without crystal structure: deep representation learning from stoichiometry. *Nat. Commun.* 11, 6280. <https://doi.org/10.1038/s41467-020-19964-7>.
41. Tshitoyan, V., Dagdelen, J., Weston, L., Dunn, A., Rong, Z., Kononova, O., Persson, K.A., Ceder, G., and Jain, A. (2019). Unsupervised word embeddings capture latent knowledge from materials science literature. *Nature* 571, 95–98. <https://doi.org/10.1038/s41586-019-1335-8>.
42. Sutton, C., Boley, M., Ghiringhelli, L.M., Rupp, M., Vreeken, J., and Scheffler, M. (2020). Identifying domains of applicability of machine learning models for materials science. *Nat. Commun.* 11, 4428. <https://doi.org/10.1038/s41467-020-17112-9>.
43. Zhou, S., Sun, Y., Gao, T., Liao, J., Zhao, S., and Cao, G. (2023). Enhanced Li<sup>+</sup> Diffusion and Lattice oxygen Stability by the High Entropy Effect in Disordered-Rocksalt Cathodes. *Angew. Chem. Int. Ed. Engl.* 62, e202311930. <https://doi.org/10.1002/anie.202311930>.
44. Ouyang, B., Artrith, N., Lun, Z., Jadidi, Z., Kitchaev, D.A., Ji, H., Urban, A., and Ceder, G. (2020). Effect of Fluorination on Lithium Transport and Short-Range Order in Disordered-Rocksalt-Type Lithium-Ion Battery Cathodes. *Adv. Energy Mater.* 10, 1903240. <https://doi.org/10.1002/aenm.201903240>.
45. Kataoka, R., Kojima, T., and Takeichi, N. (2018). Electrochemical Property of Li-Mn Cation Disordered Li-Rich  $\text{Li}^{2+}\text{MnO}_3$  with NaCl Type Structure. *J. Electrochem. Soc.* 165, A291–A296. <https://doi.org/10.1149/2.1041802jes>.
46. Ren, S., Chen, R., Maawad, E., Dolotko, O., Guda, A.A., Shapovalov, V., Wang, D., Hahn, H., and Fichtner, M. (2015). Improved Voltage and Cycling for Li<sup>+</sup> Intercalation in High-Capacity Disordered Oxyfluoride Cathodes. *Adv. Sci. (Weinh)* 2, 1500128. <https://doi.org/10.1002/advs.201500128>.
47. Nakajima, M., and Yabuuchi, N. (2017). Lithium-Excess Cation-Disordered Rocksalt-Type Oxide with Nanoscale Phase Segregation:  $\text{Li}^{1.25}\text{Nb}^{0.25}\text{V}^{0.5}\text{O}_2$ . *Chem. Mater.* 29, 6927–6935. <https://doi.org/10.1021/acs.chemmater.7b02343>.
48. Chen, R., Ren, S., Yavuz, M., Guda, A.A., Shapovalov, V., Witter, R., Fichtner, M., and Hahn, H. (2015). Li<sup>+</sup> intercalation in isostructural  $\text{Li}^{2+}\text{VO}_3$  and  $\text{Li}^{2+}\text{VO}_2\text{F}$  with  $\text{O}^{2-}$  and mixed  $\text{O}^{2-}/\text{F}^-$  anions. *Phys. Chem. Chem. Phys.* 17, 17288–17295. <https://doi.org/10.1039/c5cp02505b>.
49. Edström, K., Gustafsson, T., and Thomas, J.O. (2004). The cathode–electrolyte interface in the Li-ion battery. *Electrochim. Acta* 50, 397–403. <https://doi.org/10.1016/j.electacta.2004.03.049>.
50. Raccuglia, P., Elbert, K.C., Adler, P.D.F., Falk, C., Wenny, M.B., Molloy, A., Zeller, M., Friedler, S.A., Schrier, J., and Norquist, A.J. (2016). Machine-learning-assisted materials discovery using failed experiments. *Nature* 533, 73–76. <https://doi.org/10.1038/nature17439>.
51. Zhu, J., Wang, Y., Huang, Y., Bhushan Gopaluni, R.B., Cao, Y., Heere, M., Mühlbauer, M.J., Mereacre, L., Dai, H., Liu, X., et al. (2022). Data-driven capacity estimation of commercial lithium-ion batteries from voltage relaxation. *Nat. Commun.* 13, 2261. <https://doi.org/10.1038/s41467-022-29837-w>.
52. Roman, D., Saxena, S., Robu, V., Pecht, M., and Flynn, D. (2021). Machine learning pipeline for battery state-of-health estimation. *Nat. Mach. Intell.* 3, 447–456. <https://doi.org/10.1038/s42256-021-00312-3>.
53. Jain, A., Ong, S.P., Hautier, G., Chen, W., Richards, W.D., Dacek, S., Cholia, S., Gunter, D., Skinner, D., Ceder, G., et al. (2013). Commentary: The Materials Project: A materials genome approach to accelerating materials innovation. *APL Mater.* 1, 11002. <https://doi.org/10.1063/1.4812323>.
54. Chen, C., and Ong, S.P. (2022). A universal graph deep learning interatomic potential for the periodic table. *Nat. Comput. Sci.* 2,

- 718–728. <https://doi.org/10.1038/s43588-022-00349-3>.
55. Merchant, A., Batzner, S., Schoenholz, S.S., Aykol, M., Cheon, G., and Cubuk, E.D. (2023). Scaling deep learning for materials discovery. *Nature* **624**, 80–85. <https://doi.org/10.1038/s41586-023-06735-9>.
56. Batatia, I., Benner, P., Chiang, Y., Elena, A.M., Kovács, D.P., Riebesell, J., Advincula, X.R., Asta, M., Baldwin, W.J., Bernstein, N., et al. (2023). A foundation model for atomistic materials chemistry. Preprint at arXiv. <https://doi.org/10.48550/arXiv.2401.00096>.
57. Hong, S., Liow, C.H., Yuk, J.M., Byon, H.R., Yang, Y., Cho, E., Yeom, J., Park, G., Kang, H., Kim, S., et al. (2021). Reducing Time to Discovery: Materials and Molecular Modeling, Imaging, Informatics, and Integration. *ACS Nano* **15**, 3971–3995. <https://doi.org/10.1021/acsnano.1c00211>.
58. He, K., Zhang, X., Ren, S., and Sun, J. (2016). Deep Residual Learning for Image Recognition. In *2016 IEEE Conference on Computer Vision and Pattern Recognition (CVPR), 2016 (IEEE)*, pp. 770–778.
59. Cai, Z., Zhang, Y., Lun, Z., Ouyang, B., Gallington, L.C., Sun, Y., Hau, H., Chen, Y., Scott, M.C., and Ceder, G. (2022). Thermodynamically Driven Synthetic Optimization for Cation-Disordered Rock Salt Cathodes. *Adv. Energy Mater.* **12**, 2103923. <https://doi.org/10.1002/aenm.202103923>.
60. Kononova, O., He, T., Huo, H., Trewartha, A., Olivetti, E.A., and Ceder, G. (2021). Opportunities and challenges of text mining in materials research. *iScience* **24**, 102155. <https://doi.org/10.1016/j.isci.2021.102155>.
61. Baibakova, V., Elzouka, M., Lubner, S., Prasher, R., and Jain, A. (2022). Optical emissivity dataset of multi-material heterogeneous designs generated with automated figure extraction. *Sci. Data* **9**, 589. <https://doi.org/10.1038/s41597-022-01699-3>.
62. Szymanski, N.J., Zeng, Y., Huo, H., Bartel, C.J., Kim, H., and Ceder, G. (2021). Toward autonomous design and synthesis of novel inorganic materials. *Mater. Horiz.* **8**, 2169–2198. <https://doi.org/10.1039/d1mh00495f>.
63. Stach, E., DeCost, B., Kusne, A.G., Hatrick-Simpers, J., Brown, K.A., Reyes, K.G., Schrier, J., Billinge, S., Buonassisi, T., Foster, I., et al. (2021). Autonomous experimentation systems for materials development: A community perspective. *Matter* **4**, 2702–2726. <https://doi.org/10.1016/j.matt.2021.06.036>.
64. Szymanski, N.J., Rendy, B., Fei, Y., Kumar, R.E., He, T., Milsted, D., McDermott, M.J., Gallant, M., Cubuk, E.D., Merchant, A., et al. (2023). An autonomous laboratory for the accelerated synthesis of novel materials. *Nature* **624**, 86–91. <https://doi.org/10.1038/s41586-023-06734-w>.
65. Lunt, A.M., Fakhruddin, H., Pizzuto, G., Longley, L., White, A., Rankin, N., Clowes, R., Alston, B., Gigli, L., Day, G.M., et al. (2024). Modular, Multi-Robot Integration of Laboratories: An Autonomous Workflow for Solid-State Chemistry. *Chem. Sci.* **15**, 2456–2463. <https://doi.org/10.1039/d3sc06206f>.
66. Rohatgi, A. (2022). Webplotdigitizer. Version 4.6 (automeris.io). <https://automeris.io/WebPlotDigitizer>.
67. Paszke, A., Gross, S., Massa, F., Lerer, A., Bradbury, J., Chanan, G., Killeen, T., Lin, Z., Gimelshein, N., Antiga, L., et al. (2019). Pytorch: An imperative style, high-performance deep learning library. In *Proceedings of the 33rd International Conference on Neural Information Processing Systems*, **32**, pp. 8026–8037.