



Deep neural network battery life and voltage prediction by using data of one cycle only

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HIGHLIGHTS

- The first deep neural networks for used battery age, EoL, RUL, and cycle-by-cycle discharge voltage/power prediction.
- Highly accurate predictions based on one cycle only.
- Dimension reduction, last padding technique, and convolutional training strategy enable flexible input and high accuracy of predictions.
- Data-driven features are extracted showing more influence on battery properties than human-picked features.

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ABSTRACT

Rechargeable batteries, such as LiFePO₄/graphite cells, age differently by variability in manufacturing, charging (energy inflow) policy, temperature, discharging conditions, etc. Great economic and environmental value can be extracted if we can predict how a battery ages and ascertain its current state of health and residual useful life, based on just a few cycles of testing. Here, by developing novel-architecture deep neural networks with a special convolutional training strategy and taking advantage of recently published battery cycling data, we show that one can predict the residual life of a battery to a mean absolute percentage error of 6.46%, using only one cycle of testing. The cycle-by-cycle profiles, such as discharge voltage, capacity, and power curves of any given cycle, of used batteries with unknown age can also be accurately predicted for the first time. Moreover, our models can extract data-driven features from the data which were much more influential on the predicted properties than human-picked features. This work has shown that single cycle data contains a sufficient amount of information to predict essential battery properties with high accuracy. It is expected to provide tremendous economic and environmental benefits since reuse and recycling of batteries can be better planned and less lithium-ion batteries end up in landfills.

1. Introduction

Rechargeable batteries exemplify industrially manufactured devices. Large amounts of energy flow in and out of a battery cell, which has complex chemistry and mechanics inside, but on the outside, manifests as just current–voltage I - V , exterior temperature T , and other measurable. Battery cycle life depends on (a) the battery chemistry, (b) battery manufacturer and design, (c) variability in manufacturing, (d) history of charging and discharging, e.g. cutoff voltages, charging currents (policy), discharge (use) scenarios, ambient temperature, etc. Highly

nontrivial tradeoffs exist between the battery cycle life, the energy output (e.g. by varying the cutoff voltages [1]) and fast-charging [2]. Given a used battery of a certain manufacturer and model, accurately assessing its state of health (SOH) to predict the remaining useful life (RUL), with as little testing as possible, will provide tremendous economic and environmental benefits. It is known that by improving the battery management system (BMS) software, one could potentially double the battery cycle life [3], so less lithium-ion batteries (LIB) end up in landfills, and reuse and recycling of batteries can be better planned. In addition, as these rechargeable batteries, such as LiFePO₄ (LFP)

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cells, have been widely used in electric vehicles (EVs) [4], the resale value of used EVs can also be better estimated. Imagine one is given a used rechargeable battery of a known brand, but with an unknown number of pre-existing charge–discharge cycles s . In order to assess its SOH, one can proceed to perform a additional cycles of charge–discharge testing to acquire new $I(t)$ – $V(t)$ data. Ideally, if one can take $a = 1$, that is, with just one charge–discharge cycle testing of a used battery of unknown s , and can then predict the future cycle life (RUL) and $I(V)$ characteristics accurately, it will be tremendously helpful for building an intelligent BMS [5]. Using integer end-of-life (EoL) to denote the cycle life of a new battery just out of the factory, then $\text{EoL} = \underline{s} + a + \text{RUL}$, where underlined quantity denotes the ground truth. But since we do not know the true \underline{s} value, and thereby can only make estimates of s and RUL, there is also $\text{EoL} = s + a + \text{RUL}$, where quantities without underline are one's estimates.

The estimation and control of dynamical systems, traditionally dominated by limited-memory computations such as Kalman filter, is being challenged by deep neural network (DNN) based algorithms. While it can often be computationally intensive to train a DNN, with modern GPU/FPGA/ASIC hardware, a DNN with $\sim 10^7$ weights can be run in real-time in hand-held devices [6–8]. It thus behooves us to see how well state-of-the-art DNNs can perform in RUL–RUL for different acquisition cycling numbers a . In addition to the residual life, we could also task DNN to estimate the current age s , and by applying analysis tools like Deep Taylor Decomposition (DTD) [9], we could task the DNNs to rank the importance of *features* in reducing the root mean square error (RMSE). That is, in the acquired cycling time-sequence data $I(t)$ – $V(t)$ – $T(t)$, what might be the most important features for making accurate predictions, and how well they agree with human expert intuition.

In their ground-breaking work in 2019, Severson et al. [10] provided the world's largest open-source dataset consisting of 124 LiFePO₄/graphite cells being cycled to end-of-life EoL , defined by the cell's discharge capacity Q dropping below 80% of its out-of-factory value. Many different fast-charging policies were used on the 124 cells. EoL was shown to vary widely, ranging from 150 to 2,300 cycles, due to the variabilities (c), (d) in the beginning paragraph, with average $\langle \text{EoL} \rangle = 806$. They also proposed human-picked features based regression models to predict EoL [11], using data from the first 100 discharge cycles ($\underline{s} = 0$, $a = 100$), that is, they provided a solution to the $\text{EoL}(a = 100) \equiv 0 + 100 + \text{RUL}(\underline{s} = 0, a = 100)$ problem. Their relative mean absolute percentage error (MAPE) for EoL was 9.1% for the testing dataset, i.e. RMSE for EoL was about 167 cycles using $a = 100$. The human-picked features consist of the minimum, variance, skewness and kurtosis of the first-100 cycle discharge capacities $\{Q(m = 1 \dots 100)\}$, the slope and intercept of the linear fit to the capacity fade curve $Q(m)$ between cycles 2 to 100, the capacity at cycle 2, the difference between the maximum discharge capacity (since many types of batteries have activation) and cycle 2, the average charge time in the first 5 cycles, the integral of temperature over time in cycles 2 to 100, the minimum internal electrical resistance from cycles 2 to 100, and the difference in the internal electrical resistance between cycle 100 and cycle 2. In 2020, Attia et al. [12] utilized the same regression model to optimize closed-loop fast-charging protocols and produced more LIB cells data with even more complex fast-charging policies. Knobloch [13] developed a Convolutional Neural Network (CNN) model, applied it to the same dataset [10] and successfully reduced the required input data amount of consecutive charging cycles down to $a = 20$ while maintaining the accuracy. Tian et al. [14] proposed a DNN to predict complete charging curves by 30 points in given partial charging curves measured within 10 mins. By using the transfer learning technique, pre-trained DNNs can also be adapted to various practical scenarios, such as different ageing states, battery types, and charging strategies [15]. Yang et al. [16] proposed an extreme learning machine-based thermal model to capture the temperature behavior of batteries under external short circuit conditions, achieving better computational efficiency and accuracy than the

conventional models. Xiong et al. [17] proposed a multi-stage model fusion algorithm and a proportional–integral–differential observer, which can successfully improve the accuracy of the prediction of state of charge (SOC) and capacity under a complex application environment. Hong et al. [18] adopted the dilated CNN architecture [19] to predict RUL with RMSE of 76 based on 4 cycles ($a = 4$) of acquisition as the input data. Most of the training approaches in the literature are end-to-end, i.e. a set of specific feature correlate to a set of the target value. Thus, in the case of battery EoL prediction, it becomes difficult when only a single cycle is considered. Different from the end-to-end approach, “multi-ends-to-end” is used in the current work, i.e. multiple sets of features correlate to the same set of the target value, by the proposed last padding technique. This will be detailed in the next section.

In the present work, we show that (1) just $a = 1$ is able to provide $\text{RMSE}(\text{RUL}, \text{RUL}) < 50$, significantly better than all previous models. Furthermore, the present age s can also be predicted, with $\text{RMSE}(s, \underline{s}) < 35$. (2) The cycle-by-cycle discharge voltage curve $V(Q)$ can be predicted. This offers a great advantage for battery reuse, battery-pack modeling and EV power-system design, as connecting heterogeneous cells are the key and knowing the future $V(Q)_n$ for each cell is essential. Furthermore, the user can check the difference between cycle-by-cycle $V(Q)$ and the predicted $V(Q)$ in the future, offering confidence that the DNN is indeed working or “early-warning system” if it is not, instead of waiting till the EoL event. (3) Our DNN has identified subtle, low-dimensional battery SOH features (LDSOH) that are much sparser and superior than the human-picked features. These low-dimensional features (“latent space” in DNN) may in turn offer continuous gradient-guidelines on future battery manufacturing and usage. That is to say, since the present $\text{RUL}/\text{EoL}_{\text{DNN}}$ takes in few-cycle data to make the EoL prediction, we can visualize and “invert” our EoL_{DNN} and RUL_{DNN} efficiently by focusing on LDSOH space (e.g. 2D maps and 3D contours) and ask what the few-cycle data *should* look like in order to have better EoL_{DNN} outcome. This would allow us to ask provocative questions like, given the limited 124 cell samples of EoL ranging from 150 to 2,300 cycles, is it possible to have LiFePO₄/graphite cell with the same nameplate weight and performance rating but EoL of 3,000 cycles or even 5,000 cycles under *equally* challenging fast-charging conditions, by tweaking the fast-charging policy or even battery design? For the same battery chemistry, there are still literally hundreds of parameters in battery manufacturing, for example the electrode, separator and current collector thicknesses, calendaring pressure, electrolyte, carbon black and binder amounts, the synthesis (e.g. ball-milling, carbon coating and doping of LiFePO₄) conditions, electrolyte additives, prelithiation, formation policy, etc. So, is it possible to numerically guide the optimization of these processing and use conditions for long EoL_{DNN} and RUL_{DNN} using cheap-to-calculate and easy-to-visualize LDSOH, instead of the expensive-to-measure EoL ? Lastly, is our $\text{EoL}/\text{RUL}_{\text{DNN}}$ “explainable”? [20] (4) The architecture and training of our DNN is very flexible and generic, and should be applicable to all industrially manufactured cycling devices like washing machines, supercapacitors and material fatigue experiments. The training outcome is highly self-consistent. Indeed, examination of our RUL_{DNN} has indicated likely slight testing condition anomaly in the Severson et al. [10] dataset between the 55th to 70th testing cycles (before we noticed temperature fluctuations of the environmental chamber in their text), when presumably the testing was at its early/warm-up stage. This kind of automatic detection of the data anomaly for DNN training would allow a robust testing-and-prediction regime to be established for future cycling devices. Note that Table S1 listed n , s , m , a and all the terms used in the manuscript and the corresponding definition is added in supplementary. The illustration of n , s , m , and a are also shown in Fig. S1.

The present work aims to develop DNNs to enable higher accuracy, earlier prediction, greater interpretability and more output features to a wide range of cycling conditions [10]. Three DNNs are trained based on Severson et al.'s 2019 data of 124 LiFePO₄/graphite cells [10]. Each of

which has different main objectives, auxiliary learning tasks [21], testing strategy and purpose, summarized and listed in Table 1. The first DNN, referred to as Discharge DNN, allow the prediction of EoL, future charge time at EoL $t_{n=EoL}^{charge}$, and future voltage vs. discharge capacity $V(Q)_{n=EoL}$ curve at EoL for unseen batteries, based on cycle-by-cycle information ($m = 1 \dots a$) including human-picked features F^{human} and the “continuous- t ” data of discharge curves without the information about charging. The second DNN referred to as Full DNN, includes information about charging in the training data, and adopted a testing strategy to enable extrapolation for unknown batteries with unknown charging policies. The generality of the second DNN was significantly improved and can be applied to accurately predict EoL, cycle-by-cycle ($n = 1 \dots EoL$) $V(Q)_n$ of different batteries, even for the other dataset provided by Attia et al. in 2020 [12]. The first two DNNs can take arbitrary a from 1 to $a_{max} = 100$, but trained to assume $s = 0$ and were not tasked to predict s . The third DNN, referred to as Full RUL DNN, predicted RUL, s ($0 \leq s$), cycle-by-cycle ($n = 1 \dots EoL$) $V(Q)_n$, discharge capacity Q_n , and discharge power D_n curves for unknown age of “used” batteries. Note that Full RUL DNN went through a much larger quantity of training data since we take arbitrary starting points in the data. The proposed DNNs can accurately predict EoL/RUL and time-series properties based on an extremely small amount (even one cycle only) of input data, which can start from any cycle in the entire life of a battery. This is achieved due to the special convolutional testing and training strategy and the last padding technique adopted in the current work, giving a great flexibility of the DNNs, and in particular, the ability of detecting data anomaly. The novel deep neural networks provided highly accurate health prognostics with no prior professional knowledge of cell chemistry and the assumption of degradation mechanisms, showing remarkable predictive ability, interpretability and generality.

2. Model overview

The workflow of Discharge DNN, Full DNN, and Full RUL DNN is illustrated in Fig. 1. Note that detailed elements/building blocks/inputs/outputs etc. of each NN pipeline of all the DNNs have been illustrated in Fig. S3 to Fig. S13 in Supplementary. Here, s is defined as the number of shifted cycles from cycle number $n = 1$ used as the first cycle of the input data series, and each consisting of a consecutive cycles the testing/training strategy adopted by the DNN, which will be explained in more detail in the next section. We firstly consider the case of $s = 0$. The voltage $V(t)$, current $I(t)$, capacity $Q(t) = \int_{t_i}^{t_f} I(\tau) d\tau$, and can temperature $T(t)$ of the a consecutive cycles during the discharge process provided by Severson et al. [10] were used as input data, where $[t_i, t_f]$ are the start and finish times of the discharge half-cycle. We always discretize these continuous- t data into a vector of dimension 500 by linear interpolation, with time-step $h = (t_f - t_i)/500$, so a $4 \times 500 \times a$ “continuous- t ” data

block $x_{im}^{dis} = [V(t)_m^{dis}, I(t)_m^{dis}, Q(t)_m^{dis}, T(t)_m^{dis}]$ for each discharge half-cycle, where superscript “dis” indicates the corresponding data obtained from discharging process only; index i represents each component of the data/feature matrices; index m is defined to describe each cycle used as input and in range of $m = 1 \dots a$. Then, x_{im}^{dis} was processed by two CNNs, referred to as Dimension reduction 1 and 2, consisting of series of Inception blocks [22], residual bottleneck [23], and Luong-style attention layers [24] followed by global average and maximum pooling layers, i.e. network in network (NiN) [25] architecture. This type of model performed down-sampling by concatenating pairs of adjacent projection inputs [26]. Dimension reduction 1 and 2 gave the first estimation of EoL and charge time, denoted as \widetilde{EoL}_m^{dis} and $\left(t_{n=EoL}^{charge}\right)_m^{dis}$, based on every single cycle m during the discharge process, respectively. The two resulting $1 \times 1 \times a$ vectors from Dimension reduction 1 and 2 are the lower-fidelity version of the targets, which have great sensitivity to reflect on the variation of their target of inference. They served as “data-driven features” denoted by $F_{im}^{data,dis}$ for the networks.

In addition to the “continuous- t ” data x_{im}^{dis} which were all for discharge, we also used six human-picked cycle-by-cycle features ($6 \times 1 \times a$) denoted by F_{im}^{human} provided by Severson et al. [10] that include both charge and discharge information, for Discharge and Full DNNs. These “discrete- m ” data include (1) the Charge capacity P_m , (2) Discharge capacity Q_m , (3) Temperature average T_m^{avg} , (4) Temperature minimum T_m^{min} , (5) Temperature maximum T_m^{max} , and (6) Total charge time t_m^{charge} of each charge half-cycle m . Note that, based on our trial runs, the data of internal resistance in the dataset [10] showed the least relevance in deep Taylor decomposition (DTD) [27] analysis compared to the other human-picked features, which is somewhat counter-intuitive. Subsequently, the internal resistance feature was not used in any of our DNNs, to reduce the amount of input and save the cost of the corresponding measurements. These $6 \times 1 \times a$ “discrete- m ” features F_{im}^{human} are next

concatenated with $2 \times 1 \times a$ data-driven features $F_{im}^{data,dis} = \left[\widetilde{EoL}_m^{dis}, \left(t_{n=EoL}^{charge}\right)_m^{dis} \right]$ described in the previous paragraph, which are also

“discrete- m ” features, labeled as Discharge features (7) and (8), respectively. Our DNN is flexible with the choice of a , but we deem the practically economic maximum to be $a_{max} = 100$, which was what Severson et al. [10] used. An $8 \times 1 \times a_{max}$ data block F_{ik}^{dis} can be obtained from the $8 \times 1 \times a$ data block, by using the last-padding technique, where the last column of the $8 \times 1 \times a$ “discrete- m ” data block was copied to fully fill the fixed-width $8 \times 1 \times 100$ data block. This is the key approach to enable a great flexibility of a , and to let the same value of targets (such as EoL) being described by different lengths of input data. Then the input with more cycle information can contribute more adequate gradients for modifying the weights of the DNN and guide the training based on the input with less information, leading to an efficient learning for the DNN. The illustration is shown in Fig. S1 and the effect of this approach is detailed in Fig. S29 and 30 in supplementary.

Next the $8 \times 1 \times 100$ data block was then fed into two CNN sub-networks, referred to as Predictors 1 and 2. Predictor 1 has a structure similar to Dimension reduction with a different set of hyperparameters, and is to predict EoL and $t_{n=EoL}^{charge}$, where $t_{n=EoL}^{charge}$ prediction was an auxiliary learning task, which assisted the prediction of EoL as a hint. Predictor 2 was used to predict voltage vs. discharge capacity curves at EoL, $V(Q)_{n=EoL}$, based on the input of $8 \times 1 \times 100$ sequence matrix F_{ik}^{dis} . It adopted an attention-based encoder-decoder structure to enable this sequence-to-sequence prediction.

Full DNN further improved the generality for predicting EoL and $V(Q)_n$ curves of cells subjected to different charging policies. This was achieved by taking both charge and discharge curves into account and adopting a new design of Predictor. Similarly, Dimension reduction 3

Table 1
Summary tables for three DNNs in the current work.

Name of DNN	Main objectives	Auxiliary learning	Testing strategy	Purposes of DNN
Discharge DNN	EoL, $V(Q)_{n=EoL}$	$t_{n=EoL}^{charge}$	<1> Unselected random batteries, only trained with 100 cycles.	Predicting for unknown batteries.
Full DNN	EoL, $V(Q)_n$		<2> Unselected random charging policies, only trained with 100 cycles.	Predicting for unknown charging policies.
Full RUL DNN	RUL, Q_n , D_n , $V(Q)_n$	s	<3> Unselected random batteries, trained with 1... EoL-100 cycles.	Predicting for unknown age of unknown used batteries.

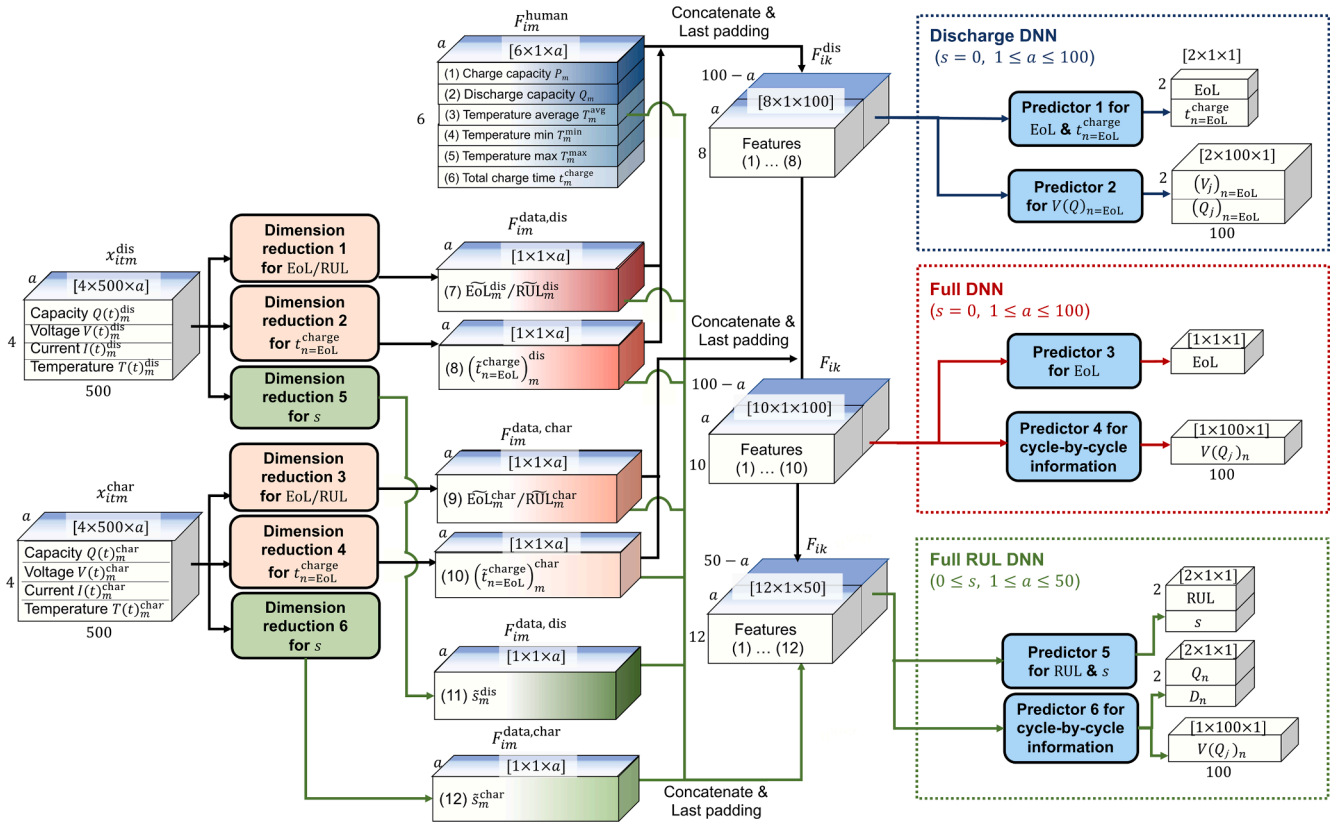


Fig. 1. The workflow of the three DNNs in the current work. Where cubes represent data in tensor form and rounded rectangles represent neural networks. The detailed elements/building blocks/inputs/outputs etc. of each NN pipeline of all the DNNs have been illustrated in Fig. S3 to Fig. S13 in Supplementary.

and 4 were introduced to extract data-driven features $F_{im}^{data, char} = \left[\tilde{EoL}_m^{char}, \left(\tilde{t}_{n=EoL}^{charge} \right)_m^{char} \right]$, which are labeled as Charge features (9) and (10) respectively, where “char” indicates the data obtained from charging process to avoid the confusion made by the symbols “charge” used by total charge time $t_{n=EoL}^{charge}$. The resulting two $1 \times 1 \times a$ vectors were concatenated with the $8 \times 1 \times a$ data block mentioned in the previous paragraphs. Last padding technique was also applied, forming $10 \times 1 \times 100$ data block F_{ik} . Next, Predictor 3 consisted of three CNN sub-networks with deep mutual learning (DML) strategy [28], and the outputted results were ensemble averaged to give the predicted EoL. In which the three sub-networks had different amounts of trainable parameters to preserve the diversity of learning. Predictor 4 provided cycle-by-cycle $V(Q_j)_n$ curves, where n can be specified by users as an integer percentage of EoL, and served as a key for the encoder part of the attention layer. It is noteworthy that Predictor 4 outputted a 1×100 vector of voltage corresponding to pre-defined Q_j ($j = 1 \dots 100$) with equally-spaced interval of $Q_{nameplate}/100$. This design reduced the complexity of the prediction of curves enabling a more efficient training/inference of model, and yet, the detail of steep changes of curves may be compromised.

Full RUL DNN has almost no limitation for positive s , i.e. the input sequence does not necessarily begin from the cells first cycle. However, $1 \leq a \leq 50$ was set to reduce the extremely large amount of data in training process due to the consideration of the wide range of s and a . Therefore, instead of EoL, RUL can be predicted. This also indicates that the networks can adapt and respond correctly based on the features in any cycle as its first input. Full RUL DNN aims at truly well used battery cells, that may have $\bar{s} > EoL/2$ already. Full RUL DNN required two additional Dimension reduction 5 and 6, extracting the data-driven features \tilde{s}_m^{dis} and \tilde{s}_m^{char} , labeled as (11) and (12), respectively. Thus, in

total 12 features of a cycles started from the unknown s^{th} cycle can be obtained.

In addition to RUL, cycle-by-cycle Q_n , D_n , and $V(Q_j)_n$, the value of s was also predicted by Full RUL DNN as an auxiliary learning task. Thus, Full RUL DNN can output the predicted current age s of any well-used batteries, which can be a validation check for any user who doubt the efficacy of the DNN (“ask the Fortune Teller my age, before trusting her prediction of the Doomsday”). A special “convolutional training” strategy was applied to both Full RUL DNN to deal with such complicated tasks where both s and a are varying with little constraint.

2.1. Network setting and training

Our DNNs used the unmodified “raw data” provided by Severson et al. [10] Certain data were, however, omitted due to their unsatisfactory quality specified in the exception notices in the experiment log, such as early stoppage of the tests, temperature recording failure, or data with excess noises. In total, 115 batteries with an acceptable quality of discharge data in the first 100 cycles were considered in our Discharge DNN; out of these, 95 batteries were considered in Full DNN as the charge curves data quality of the omitted 20 batteries were not of satisfaction. Meanwhile, only 81 batteries have acceptable data quality in all the discharge and charge curves in every $0 \leq s, m = 1 \dots a$ cycle, and thus can be considered in Full RUL DNN. Here the constant-current regions of the discharge curves, and the entire curves of charging process of capacity, voltage, current, and temperature were utilized as input datasets.

Three testing strategies were adopted corresponding to the three DNNs proposed in our work. Discharge DNN was designed for the prediction of unlearned battery cells. Thus, its testing strategy, referred to as testing strategy $<1>$, was that the data of entire sequences of certain randomly chosen batteries was omitted from all the considered

batteries. The resulting datasets then trained and forced Discharge DNN to learn the features of overall sequences and to extrapolate for different batteries. Next, Full DNN was designed for batteries with varying charging policies. Testing strategy < 2 >, where the data of entire sequences of all the batteries with randomly chosen charging policies were deliberately omitted, were used by Full DNN to train the ability of extrapolation for different charging policies. The training of the two DNNs mentioned above was trained by the first 100 cycles ($s = 0$, $1 \leq a \leq 100$) of each battery only, i.e. the information of the remaining cycles ($a > 100$) was not considered. In contrast, Full RUL DNN adopted a special “convolutional training/testing” (CT) strategy < 3 >, where the starting cycle with s shifted cycle from $n = 1$ and the number of cycles used a in each set of training data may vary. The kernels with all the possible combinations of s and a then convoluted throughout the entire life cycles of 65 randomly selected batteries (around 80% of the 81 considered batteries) in the training set. This allows Full RUL DNN to possess the ability of multi-vision to obtain full information about the entire life of batteries. Thus, accurately predicting RUL based on an arbitrary s with little information of a cycles can be achieved. The detail of the training strategies < 1 >...< 3 > was illustrated in Fig. S14 in Supplementary.

2.2. Performance of discharge and Full DNNs

The performance of Discharge and Full DNNs is shown in Fig. 2. There are 115 cases for Discharge DNN and 95 cases for Full DNN in the figures, i.e. the total number of batteries considered in the DNNs. The mean absolute percentage error (MAPE) of the predicted values to ground truth of EoL corresponding to the training and testing sets were shown in the legend of each diagram in Fig. 2(a). It is remarkable that very low testing MAPE(EoL, EoL₀) = 6.46% can be achieved by Discharge DNN based on $F_{ik}^{\text{discharge}}$, i.e. the discharge curves and the six human-picked features, at the first cycle only ($a = 1$). Full DNN can also give low testing MAPE(EoL, EoL₀) = 7.78% for the predicted EoL. These MAPE values based on one cycle only are even lower than 9.1% based on 100 cycles in the literature [10]. Our testing MAPE of Full DNN can be further improved to lower than 3.97% when the data of the first 100 consecutive cycles were used ($a = 100$), but we consider such improvement in quality to be disproportional to the $100 \times$ input measurement work required. In addition to such high overall accuracy of prediction, sparse data points and outlier cases with extremely low or high lifetimes can still be captured by both the DNNs.

Discharge DNN can predict $V(Q)_{n=EoL}$ curve. Two batteries with the least and the greatest differences on curves at $n = 1$ and EoL , i.e. the best

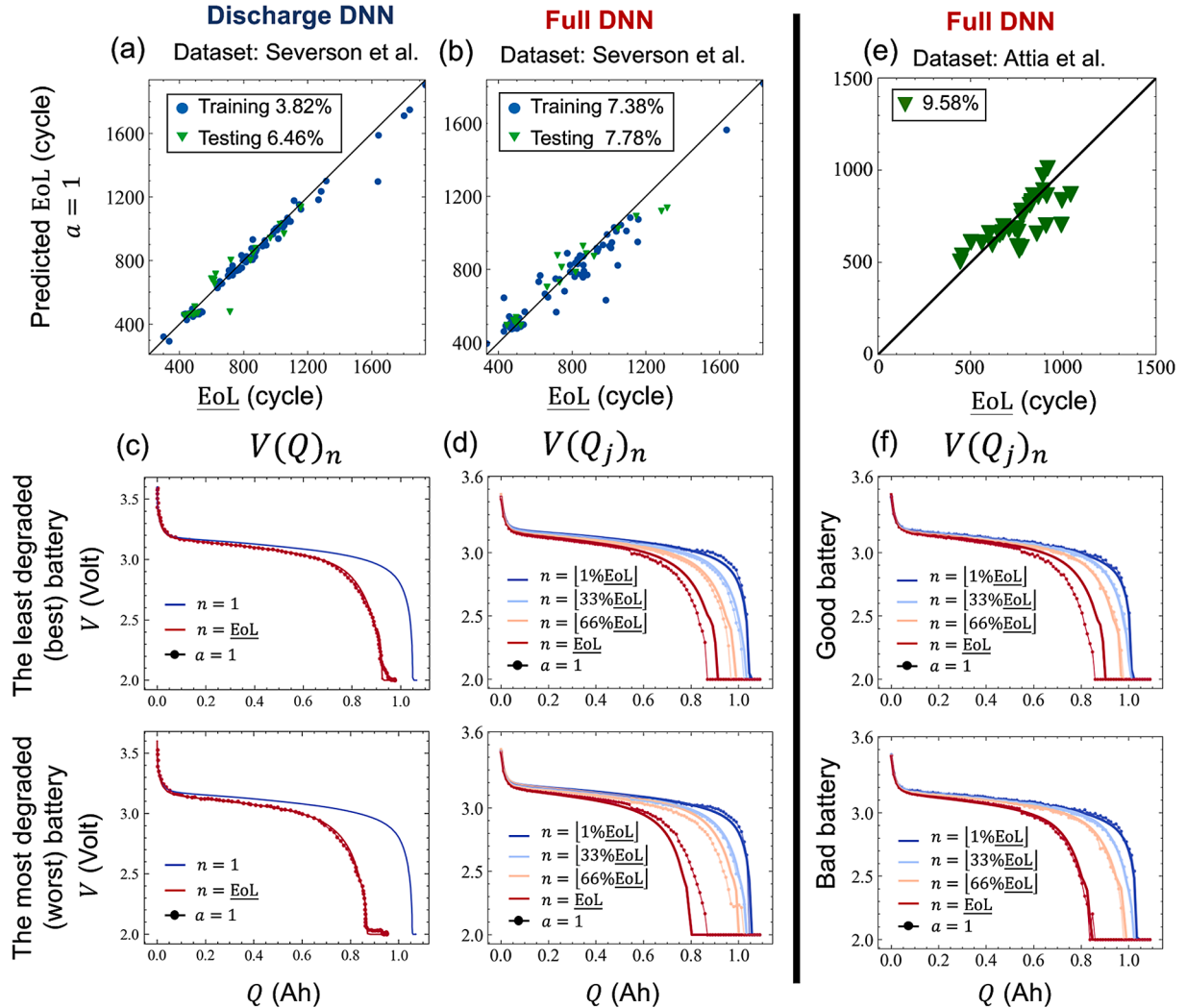


Fig. 2. (a)(b) The predicted values EoL vs. EoL_0 of Discharge and Full DNNs based on the first cycle ($s = 0, a = 1$) in data of Severson et al. Low MAPE (<7.78%) were achieved. (c)(d) The predicted $V(Q)_n$ curves by the two DNNs for the cases of the least degraded (best) and the most degraded (worst) batteries. In the second part of the figure, Full DNN is applied to predict (e) EoL of 34 cells provided by Attia et al. [12] with totally different charging policies, and (f) $V(Q)_n$ curves of two of batteries in (e) are also predicted.

and worst batteries in the dataset, are shown in Fig. 2(c) as examples. Here the difference in $V(Q)_{n=\text{EoL}}$ was defined by the integrated area between the two curves at $n = 1$ (blue solid line) and EoL (red solid line), and can be regarded as an indicator of the level of ageing. The dotted line was the degraded curve predicted based on the first ($a = 1$) cycle, while the red solid line was the ground truth of curves at EoL . It can be observed that the dotted and red solid lines are almost identical, showing a great predictive power of our DNN in both long-lived and short-lived outliers.

We next examine the performance of Full DNN on the prediction of cycle-by-cycle $V(Q)_n$ curves, as shown in Fig. 2(d). Where solid lines in blue, light blue, orange, and red representing the ground truth of curves of $n = 1\%[\text{EoL}]$, $33\%[\text{EoL}]$, $66\%[\text{EoL}]$, $[\text{EoL}]$ are chosen for illustration purposes, where $[x]$ is floor operation giving the greatest integer less than x . It can be observed that Full DNN accurately predicted these curves and successfully captured the history of the ageing process of cells. Note that our DNN provided a more accurate prediction on the curves for the early-stage usage of cells, since cells in their early stage typically are at a relatively healthy state and with less uncertainty.

The error metrics of properties predicted by Discharge and Full DNNs were shown in Table 2. It is remarkable that, with the average $\langle \text{EoL} \rangle = 806$ cycles and a standard deviation of 377 in the batteries, RMSE of 57 cycles for EoL prediction can be achieved by Discharge DNN based on the first-cycle data only ($s = 0$, $a = 1$). It is significantly less than the benchmark of RMSE of 122 obtained from Severson et al. based on the first 100 consecutive cycles ($s = 0$, $a = 100$). RMSE was dramatically dropped to 33 when data of the first 100 consecutive cycles were considered in Discharge DNN. In Full DNN, RMSE of 58 and 39 cycles for EoL prediction can be achieved based on data of the first ($a = 1$) and the first 100 cycles ($a = 100$). The ability of extrapolation of Full DNN was excellent and illustrated in Fig. S15 in Supplementary. It is of interest that Full DNN and Discharge DNN produced very similar RMSE. This shows that the auxiliary learning task for predicting future charge time $t_{n=\text{EoL}}^{\text{charge}}$ in Discharge DNN was very helpful in predicting EoL. The effect of this design was almost equivalent to the input data of the entire charge part for Full DNN.

The error metric \mathcal{M}_n for $V(Q)_n$ curves predicted by Discharge DNNs was also shown in Table 2. It is defined by

$$\mathcal{M}_n = \frac{\sqrt{\frac{1}{100} \sum_{j=1}^{100} \left((\Delta V_{jn} / (V^{\text{upper}} - V^{\text{lower}}))^2 + (\Delta Q_{jn} / Q_{\text{nameplate}})^2 \right)}}{\int |V'(q)|_n dq}$$

Table 2

Comparison of error metric of properties predicted by the current DNNs and Severson et al. [10]. The superscript < > indicates the specific training strategy adopted by the DNN.

Situation	First cycle $s = 0$ $a = 1$	First 5 cycles $s = 0$ $a = 5$	First 100 cycles $s = 0$ $a = 100$	First 100 cycles $s = 0$ $a = 100$
	Discharge DNN			Discharge Model in [10]
RMSE(EoL, EoL_-) (cycles)	44/57	38/49	15/33	76/122
RMSE($t_{n=\text{EoL}}^{\text{charge}}, t_{n=\text{EoL}}^{\text{charge}}$) (mins)	0.37/ 0.6	0.38/ 0.53	0.17/0.45	–
$\mathcal{M}_{n=\text{EoL}}$	32/43	23/36	17/16	–
	Full DNN			Full Model in [10]
RMSE(EoL, EoL_-) (cycles)	50/58	42/48	35/39	51/167
$\langle \mathcal{M}_n \rangle$	15/26	12/20	10/22	–

where ΔV_{jn} and ΔQ_{jn} are the difference between the j^{th} data point of ground truth and predicted values of voltage and charge for the given cycle n ; $[V^{\text{lower}}, V^{\text{upper}}]$ is the discharging and charging voltage cutoffs; $Q_{\text{nameplate}} = 1.1$ Ah, i.e. the nameplate discharge capacity of these 18,650 LiFePO₄/graphite cells; the denominator is the total length of curve. For the prediction of cycle-by-cycle $V(Q)_n$ curves by Full DNN, the error metric, $\langle \mathcal{M}_n \rangle$, is determined by averaging $\mathcal{M}_{n=1\% \text{EoL}}$ through $\mathcal{M}_{n=\text{EoL}}$ for all the 100 $V(Q)_n$ curves. It is remarkable that even though the complexity of the prediction of cycle-by-cycle curves is significantly higher than it of the prediction of a single-cycle curve at EoL, their error metric values listed in Table 2 are very similar to those of Discharge DNN.

Overall, the error metrics difference between testing and training sets in all items of Table 2 was converged when the first 100 consecutive cycles were considered. This indicated that the feature learning process of the three DNNs were all in good condition and without obvious overfitting phenomenon. The learning progress of the current DNNs was illustrated in detail in Supplementary Fig. S16...18. The reproducibility and reliability of the Discharge and Full DNNs were examined by five-fold cross-validation tests. Five different training sets were used to train the two DNNs. The RMSE of EoL curves show slight fluctuations in the early stage and quickly converge after input cycle $a > 30$ showing high robustness, as shown in Fig. S19 in Supplementary.

With the advantage of highly accurate prediction, our DNNs are particularly suitable for closed-loop optimization for maximizing battery cycle life recently proposed by Attia et al. [12]. As RMSE of our EoL_{DNN} ($a = 1$) was even less than the benchmark which used $a = 100$ cycles [10], as shown in Table 2, the required measuring number of cycles and optimization time in the protocols can be significantly reduced. Moreover, apart from accurately predicting EoL, our Discharge DNN can predict additional properties with very high accuracy, such as the total charge time. This also enables a useful extension for the protocols to optimize EoL and charge time simultaneously to achieve different objectives defined by the users who may favor the maximum EoL, the shortest charging time, or any preferable combination of the two.

2.3. Application to batteries with different charging policies

The generality of the current work was examined by applying Full DNN based on the year-2019 dataset to predict EoL of batteries included in a completely different dataset which is provided by Attia et al. [12] in 2020, without retraining. The batteries were subjected to very complex charging policies with 5 different constant-current steps. There were 34 batteries chosen from the 2020 dataset [12] to test our Full DNN. Note that 11 batteries in the dataset were omitted since the data condition of their first cycle does not have acceptable quality for the inference of EoL. For example, their current curves are greatly shifted; the patterns of their temperature curves are different from most of the batteries in the dataset. The corresponding data screening process is detailed in the supplementary. Fig. 2(e) shows that Full DNN has MAPE(EoL, EoL_-) = 9.58% based on the first cycle only ($a = 1$), which is slightly higher than those when Full DNN predicted EoL in the testing dataset provided by Severson et al. [10]. We believe this is because the significant difference in the discharge capacity of batteries used in the Attia et al. [12] (1.051 ± 0.003 Ah) and Severson et al. [10] (1.078 ± 0.002 Ah). Even though, MAPE of 9.58% can still be regarded as sufficiently low for the data and charging policies which were not learned by the DNN. More detail can be found in Fig. S21 and S22 in supplementary.

We next examined the performance of Full DNN in predicting time-series properties $V(Q)_n$ of the good and bad batteries. Note that the curves predicted by Full DNN required two automatic pre-processing steps for illustration purposes, as the dataset provided by Attia et al. [12] was not known to the DNN. First, a rigid shift should be performed to make the first point of the predicted curve matches that of the initial

curve of the first cycle (i.e. the voltage at zero discharge capacity). After the rigid shift, the second process was to omit the points in the predicted curve with voltage lower than the minimum applied voltage specified in the dataset (2.0 V in this case). This data pre-processing is completely general and tamper-proof, as all the information required here was included in the input data of the DNN. Fig. 2(f) shows that Full DNN successfully predicted the curves with high accuracy for both cases. It is of interest that, for the case of the bad battery, the ground truth (solid lines) and predicted (dotted lines) curves were almost identical, while there was a notable difference for $n = \text{EoL}$ in the case of the good battery. This is because the degradation was insignificant in the good battery, and fewer aging features can be discovered by the DNN in the first cycle. Similar conclusions were made in the work of Zheng [29], where the incremental open circuit voltage test for better estimation of SOC are recommended.

2.4. The most influential features for predicting EoL

The network design enables the interpretation of the current DNNs. The total counts for the human-picked and data-driven features on EoL being the most influential feature in each input cycle in all batteries in the two DNNs can be revealed by Deep Taylor decomposition (DTD), as shown in Fig. 3. There are 115×100 and 95×100 counts in total in Fig. 3(a) and (b), respectively. In Discharge DNN (Fig. 3(a)), the top 3 influential features were data-driven features (7) $\widetilde{\text{EoL}}_m^{\text{dis}}$, (8) $\left(\widetilde{t}_{n=\text{EoL}}^{\text{charge}}\right)_m^{\text{dis}}$, and human-picked feature (5) Temperature maximum T_m^{max} . In Full DNN, the top 3 influential features were data-driven features (10) $\left(\widetilde{t}_{n=\text{EoL}}^{\text{charge}}\right)_m^{\text{char}}$, (8) $\left(\widetilde{t}_{n=\text{EoL}}^{\text{charge}}\right)_m^{\text{dis}}$, and human-picked feature (5) Temperature maximum T_m^{max} . In contrast, the feature with relatively low relevance in both the DNNs was human-picked feature (1) Charge capacity P_m . Remarkably, the results showed that the most influential features to EoL of a battery are all data-driven features extracted by our Dimension reduction, rather than the human-picked features suggested by the literature, by as much as one order of magnitude. This demonstrated the power of DNNs which is capable of discovering the crucial underlying factors for the performance over and above human intuitions.

We next visualized the most influential feature for each of 95 batteries with different EoL in Full DNN, as shown in Fig. 3(c). It can be observed that data-driven features are more relevant than other features to the batteries with long-range of EoL, showing their great applicability to batteries with different qualities. Furthermore, it is of interest to note that Charge data-driven features (9) $\widetilde{\text{EoL}}_m^{\text{char}}$ and (10) $\left(\widetilde{t}_{n=\text{EoL}}^{\text{charge}}\right)_m^{\text{char}}$ are more relevant to the batteries with EoL below the average value of 806 cycles (marked by a dashed line) while Discharge data-driven features (8) $\left(\widetilde{t}_{n=\text{EoL}}^{\text{charge}}\right)_m^{\text{dis}}$ are more relevant to those with longer EoL. This result

implies that charging policies are the main reason to cause aging and degradation of batteries. It is also worth noting that, batteries with EoL around the average value typically behaved neutrally without significant characteristics. Thus, it is reasonable that both human-picked and data-driven features were possibly the most influential ones. In our Supplementary, more correlations between EoL and input data were determined by layer-wise relevance propagation (LRP) method in Fig. S23, and selected parts of input data suggested by LRP were also visualized and enlarged in Fig. S24.

2.5. The prediction of RUL and the current battery age s

Now we examine the performance of RUL prediction by Full RUL DNN, where $0 \leq s, 1 \leq a \leq 50$. Fig. 4(a) and (b) show the testing RMSE (RUL, $\widetilde{\text{RUL}}$) and RMSE(s, \underline{s}), respectively, where s can be regarded as the unknown age of a used battery. Since this DNN allows accept input from wide range of s (1–1684 cycles), three representative ranges corresponding to “Baby” age (1–100 cycles), “Mid” age (401–500 cycles), and “Old” age (701–800 cycles) were chosen for illustration purpose. RMSE diagrams for RUL and s in the full range are shown in Fig. S25 in Supplementary. It can be observed that RUL was accurately predicted by Full RUL DNN. Most of the regions in Fig. 4(a) are in blue, i.e. RMSE (RUL, $\widetilde{\text{RUL}}$) < 50.

It is of interest that, in Fig. 4, the regions with relatively high RMSE form 45° sharp lines, i.e. $a + s_- = c$, where c is a constant. This is because the last padding technique used here repeated the information of the last input cycle, which was equivalent to assign a greater weight to that cycle in the DNNs. In other words, whenever the data of a single cycle with low quality of measurements was the last element in the input data, the prediction was then greatly misled by that particular cycle, resulting in a sudden increase of RMSE. Similarly, RMSE may also drop immediately when the last input element was with an acceptable quality. For example, region A in Fig. 4(a) with relatively high RMSE is a 45° band located around $s_- = 55$. It is because an anomaly condition (temperature fluctuations of the environmental chamber according to Severson et al. [10]) occurred around those cycles for half of the batteries in the dataset. This indicates that the current DNN successfully reflects the anomaly conditions that occurred in the testing of given batteries. In addition, even the input was with such anomaly conditions, RMSE of the current DNN is still lower than 50 cycles, and it can be further reduced by simply considering more input cycles (greater a). This great flexibility, reliability and robustness are not offered by any other DNN models in the literature which only accept fix amount of input cycles. Another region with relatively high RMSE is $s_- \leq 15, a \leq 15$ marked by triangle B. It is reasonable as the predictions were based on small amount of data and batteries were typically at a relatively healthy state with almost no aging features in their early/warm-up stage. It is worth noting that the first column ($s_- = 0$) in Fig. 4(a) can be compared with the results of Full DNN shown in Table 2. Where their RMSE were both in a similar range between 40 and 80 cycles, even the prediction of RUL is more complex than that of EoL as \underline{s} is unknown and

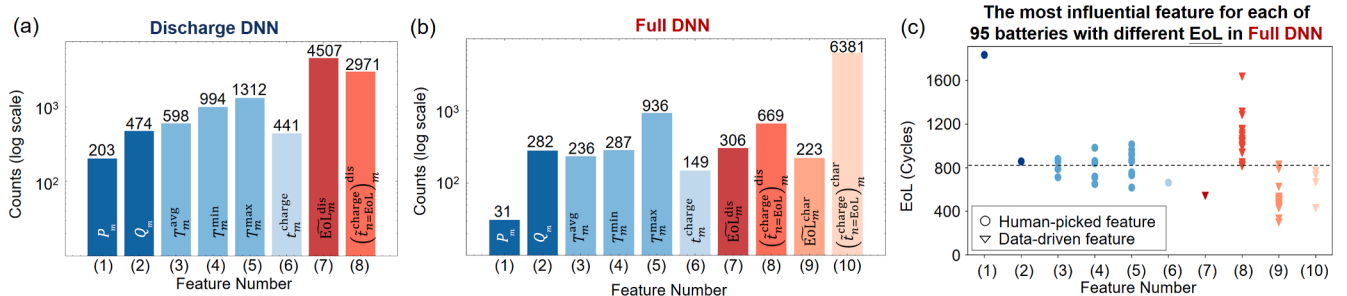


Fig. 3. The total counts for the human-picked and data-driven features on EoL prediction by (a) Discharge and (b) Full DNNs being the most influential feature in each input cycle in all batteries, obtained by Deep Taylor decomposition. (c) The most influential feature for each of the 95 batteries with different EoL in Full DNN.

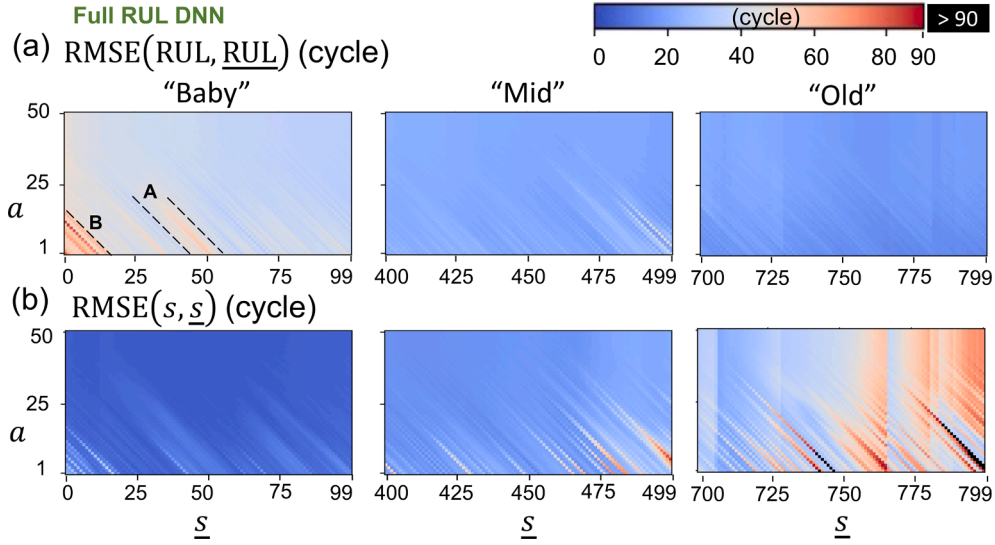


Fig. 4. Testing RMSE of predicted (a) RUL and (b) s , which can be regarded as the current battery age, by Full RUL DNN.

flexible in Full RUL DNN.

Fig. 4(b) shows the performance of age prediction s by Full RUL DNN. It can be observed that the occurrence of 45° sharp edges with relatively high RMSE had good agreement with those in Fig. 4(a), showing a high correlation between RUL and s . Full RUL DNN typically provided a more accurate prediction of s when s is small, as shown in Fig. 4(b). In most of the cases, low RMSE(s, s) (<35 cycles) can be achieved when $s < 750$. However, RMSE(s, s) went up as $s > 750$ (see Fig. S25 in Supplementary for more detail). This is because the amount of training data corresponding to the batteries with $\text{EoL} > 750$ cycles reduced to 17.3% of the entire dataset (see Fig. S26 in Supplementary), providing insufficient information for Full RUL DNN to learn the corresponding features.

We next examine the performance of Full RUL DNN on cycle-by-cycle information. Fig. 5(a)(b) shows testing MAPE of discharge power D_n and discharge capacity Q_n values at the n^{th} cycle with $0 \leq s_-, a = 1$. It is remarkable that our MAPE is typically <4% based on the data at any

values of s , showing excellent cycle-by-cycle predictability. However, D_n and Q_n were relatively difficult to be predicted when n was close to EoL as the batteries became unstable with higher uncertainty, giving a thin black band region at the top of the figures. It is also of interest noting that, for both D_n and Q_n , high accuracy can be achieved especially when the values of the predicting target cycle n and the input data age s were close, resulting in a dark blue band from bottom left to top right in Fig. 5(a)(b). These results were also reasonable as the less cycle difference between n and s , less uncertainty may be introduced in the batteries. Fig. 5(c) is the error metric \mathcal{M}_n for $V(Q)_n$ curves, showing very low values (typically $\mathcal{M}_n < 4\%$). However, a thin black band region at the top of the figures still existed for the same reason mentioned above. A relatively higher value of \mathcal{M}_n close to 4% can be found in the left half of Fig. 5(c). This is because the batteries at their early-stage (less s) were still healthy with less ageing information. In contrast, age left its traces

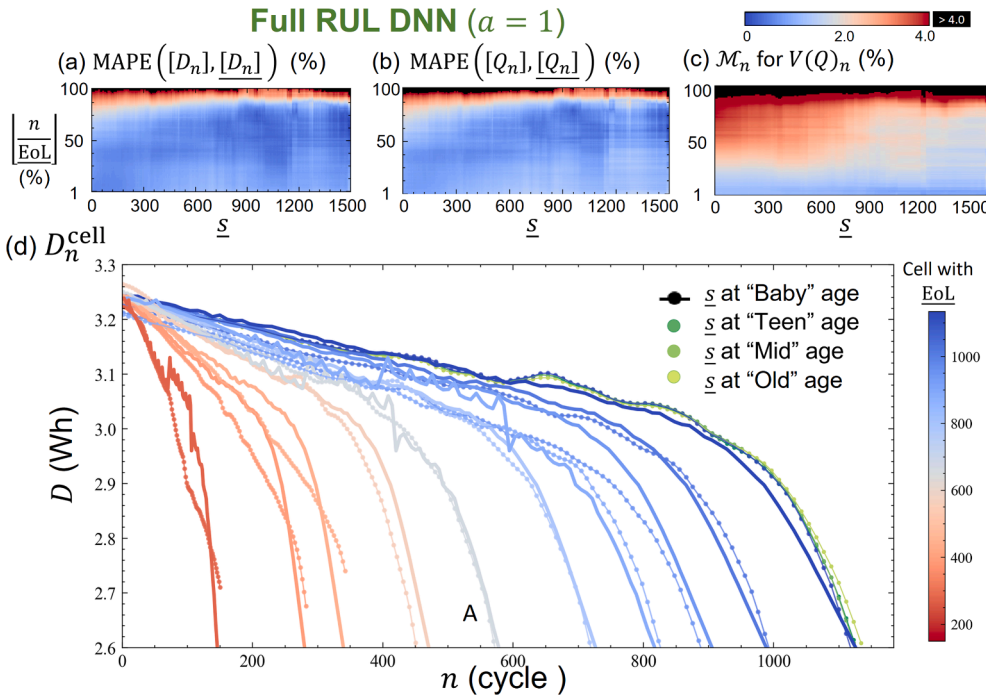


Fig. 5. Testing MAPE of (a) discharge power D_n , (b) discharge capacity Q_n and (c) the error metric \mathcal{M}_n for $V(Q)_n$ predicted by Full RUL DNN with $0 \leq s_-, a = 1$. (d) The ground truth of discharge power curves D_n^{cell} of ten cells with various EoL provided by Severson et al. [10] and the predicted dotted curves by Full RUL DNN based on one cycle only ($a = 1$) at their "Baby" age ($s_- = 0$). The predicted green dotted curves based on "Teen," "Mid," and "Old" ages, i.e. $[(\text{EoL} - 100) \times 33\%]$, $[(\text{EoL} - 100) \times 66\%]$, $\text{EoL} - 100$, are also shown for the longest EoL battery.

on the data at the later stage (greater s), providing sufficient information for our DNN to predict $V(Q_j)_n$ curves accurately in a wide range of n .

Fig. 5(d) shows the predicted (dotted lines) and ground truth (solid lines) of discharge power curves D_n^{cell} of ten selected batteries with various EoL provided by Severson et al. [10]. The prediction was performed based on one cycle only ($a = 1$) at the age of “Baby” ($s_- = 0$). It is remarkable that several important features of the curves of all the ten curves were accurately predicted. For example, the endpoints of most of the predicted curves where $D = 2.6$ Wh matched well with those of ground truth; slope changes of the curves were also accurately predicted; some of the batteries (e.g. battery A marked in Fig. 5(d)) had special fluctuation before the final steepest drop and our DNN can reveal this phenomenon in the predicted curve. We also plot three green dotted predicted D_n^{cell} curves for the batteries with the longest EoL as an example, based on the input cycles of “Teen,” “Mid,” and “Old” ages (i.e. $\lfloor (\text{EoL} - 100) \times 33\% \rfloor$, $\lfloor (\text{EoL} - 100) \times 66\% \rfloor$, $\text{EoL} - 100$), respectively. Note that, the last 100 cycles before EoL were omitted here, as these data were relatively unstable and may have uncommon features of batteries. It can be observed that there was no significant difference between the predicted D_n^{cell} curves based on the four ages of s as input, demonstrating the robustness of the current DNN.

Overall, Full RUL DNN was proven to be a powerful tool to accurately predict RUL, age, and cycle-by-cycle information of used batteries based on input data of few consecutive a cycles started from any cycle s . The cycles under anomaly conditions can also be identified. Most importantly, the results proved that the amount of required data for accurate RUL prediction for any given battery can typically be as small as one cycle ($a = 1$) and including more input cycles ($a > 1$) has limited effect on improving the prediction accuracy. We also show that the data of each cycle of the battery contains sufficient amount of information to be distinguished from each other, provided that the level of precision/resolution of the measurement is sufficiently high in Fig. S27 in the supplementary.

3. Discussion

The proposed DNNs have several advantages compared with the traditional key index measurement methods in practical application. First, a more efficient, convenient, and practicable data storage can be achieved due to the two following reasons. (1) Only a single cycle of data is needed for our DNNs to achieve highly accurate prediction and (2) the stored data of that single cycle can be any cycle before EoL for our Full RUL DNN. This is a significant improvement compared with the conventional machine learning BMSs, which typically required saving the whole sequence of many cycles to predict EoL . Second, take the key index $\Delta Q_{100-10}(V)$ suggested by Severson et al. [10] as an example. This index shows a very high correlation with EoL of battery, and yet, requires long measurement, i.e. the 10th and 100th cycles. On the other hand, with the help of Dimension Reduction and last padding technique, our DNNs require measurement on any given single cycle and significantly reduce time and cost. Third, the proposed DNNs is suitable to constantly adjust the model to deal with the interference of environmental change and improve prediction accuracy in practical applications. It is because our DNNs are constructed by two parts: Dimension Reduction and Predictor as shown in Fig. 1, offer training flexibility that the users can choose one of the part or both for adjusting the weights.

As for potential applications for lithium-ion batteries, it is possible that our DNNs can be used as the early warning and the safety performance system. For example, since our DNNs can predict the future cycle-by-cycle information (such as $V(Q_j)_n$ and D_n^{cell} curves for the future n -th cycle) based on any previous single cycle of a given battery, these predicted curves can serve as baseline for the future usage of that battery. The users can keep using that battery until the n -th cycle, and compare the actual curves with the predicted base line curves of the n -th cycle. If

the actual curves appear worse than predicted baseline curves, it means that the battery has been damaged by unexpected conditions. In this way, the early warning about the health status of that battery can be achieved. Moreover, our DNNs can serve as the predictor in the system of high-throughput battery fabrication, testing, screening and optimization. For example, our DNNs can instantly screen out the just-manufactured batteries with unacceptable predicted EoL ; developers can also use our DNNs to predict EoL of batteries subjected to different charging strategies, and thus, charging strategy can be optimized. This approach can also be applied to the optimization of manufacturing parameters.

In this work, we have shown that single cycle data contains a sufficient amount of information to be distinguished from each other and can be used to predict EoL , RUL and other related properties with high accuracy, even for a completely different dataset (Attia et al. 2020 [12]) without retraining. However, we still need to emphasize that the datasets applied in the current work are measured in laboratories. Our DNNs may need more training data to adapt more complex charging strategies, wider engineering/experience of practical aspects, and broader device type/chemistry.

4. Conclusion

This paper first presents two deep neural networks (DNNs) with a novel network design to predict End-of-Life (EoL), charge time t^{charge} , and discharge voltage curve $V(Q)$ curves at EoL of given batteries. To our knowledge, this is the first work to achieve testing root mean square error (RMSE) of EoL prediction as low as 57 and 33 cycles based on inputs of the first cycle ($a = 1$) and the first 100 cycles ($a = 100$) only, respectively (where $\langle \text{EoL} \rangle = 806$ cycles in the dataset). This was attributed to the introduction of Dimension reduction which extracted data-driven features from the measured profiles of input properties, and the last padding technique. Our results show that these data-driven features played more important roles and were much more influential on the predicted properties in the inference of the DNNs, compared to human-picked features such as the charge/discharge capacity (P_m , Q_m), and the maximum/minimum temperature (T_m^{max} , T_m^{min}). Moreover, our Full DNN was trained to focus on the overall sequences of properties in each cycle with different charging policies, giving very low RMSE of predicted EoL even for batteries with unknown charging policies. The generality of the current DNN was also examined by applying to a completely different dataset, giving the prediction of EoL and cycle-by-cycle information, such as discharge capacity Q_n , discharge power D_n and $V(Q_j)_n$ curves at any given n -th cycle with very high accuracy.

We also proposed a powerful Full RUL DNN which allows the prediction of remaining useful life (RUL) and the current battery age s , and most importantly, allow the flexibility of both s and a . The last padding technique during the training process enables such flexibility, strengthening the connection of the underlying time-series features between sections of data with different length. Also, the performance of the current networks improved as the amount of training data increases, showing a healthy condition of the networks. The anomaly environmental conditions during the tests of batteries can also be detected in the proposed network.

With the help of the proposed special convolutional training strategy throughout the entire life cycles, the DNN enables a more accurate prediction ($\text{RMSE}(\text{RUL}, \text{RUL}) < 50$ cycles in the most of the range of cycles), a more accurate cycle-by-cycle information prediction (error metrics $< 4\%$ for the predicted properties), a much smaller amount of required cycles of input data (typically only 1 cycle is needed), and more flexible length of input data (s and a can be arbitrary) than the state-of-the-art algorithm. Thus, practically, our key contributions, including high accuracy of prediction, high efficiency on saving battery history and excellent ability of extrapolation for unknown charging policies and unknown age of used batteries, make our DNNs suitable to be an

important component of modern quality control systems in battery manufacturing and battery management systems. The current work demonstrated the power of machine learning, which sheds light on solving the long-lasting problems of health estimation and lifetime prediction of lithium-ion batteries, providing great economic and environmental benefits.

4.1. Methods

In the current work, all the data sets were standardized, such that

$$Z \equiv \frac{x - \mu}{\sigma}$$

where Z is standardized value; x is the original value from the raw data; μ and σ represent the mean and standard deviation of the raw data, respectively. These standardized data served as the training and testing data sets for the all DNNs. The performance of the DNNs was evaluated by the root mean square error (RMSE), such that

$$\text{RMSE} \equiv \sqrt{\frac{1}{n} \sum_{i=1}^n (y_i - \hat{y}_i)^2}$$

where n is the total amount of sample; y_i and \hat{y}_i are the ground truth and predicted values, respectively. All the DNNs achieved relatively lower RMSE than other competitors, showing that key features in the profiles of the properties were successfully captured and correlated with EoL/RUL of batteries. Mean absolute percentage error (MAPE) also served as an error metric for cycle-by-cycle information, e.g. Q_n and D_n , such that

$$\text{MAPE} \equiv \frac{1}{n} \sum_{i=1}^n \left| \frac{y_i - \hat{y}_i}{y_i} \right|$$

We also used Huber loss [30] during the training process for predicting $V(Q_j)_n$ curves, Q_n and D_n , such that

$$\text{Huberloss} \equiv \begin{cases} \frac{1}{2}(y_i - \hat{y}_i)^2, & \text{if } |y_i - \hat{y}_i| < \delta \\ \delta|y_i - \hat{y}_i| - \frac{1}{2}\delta^2, & \text{otherwise} \end{cases}$$

where δ is a hyperparameter which makes the behavior of loss function similar to MSE or MAE.

We also applied Deep Taylor decomposition (DTD) [9] to the DNNs to enable the feature explanation. It redistributed the relevance layer by layer from the predicted results back to the input data. This analysis provided the most influential key feature on target, showing that the data-driven features obtained by Dimension reduction were, in general, more important than human-picked features.

Moreover, in order to obtain the attribution along with the profile of input data, relevance score R_j for the j -th neuron in each layer based on the k -th neuron in the next layer was calculated by using either LRP- $\alpha\beta$ or LRP- ϵ methods according to the type of the present layer (convolution or dense layers, respectively). These relevance scores were then reweighted by the flat method to have their summation as a unity. The formulation of LRP- $\alpha\beta$ and LRP- ϵ method can be expressed as

$$R_j = \sum_k \left(\alpha \frac{(a_j \omega_{jk})^+}{\sum_{0,j} (a_j \omega_{jk})^+} - \beta \frac{(a_j \omega_{jk})^-}{\sum_{0,j} (a_j \omega_{jk})^-} \right) R_k$$

$$R_j = \sum_k \left(\frac{a_j \omega_{jk}}{\epsilon + \sum_{0,j} a_j \omega_{jk}} \right) R_k$$

where a and ω are the output and weight of the current layer; the notation $(x)^+$ and $(x)^-$ are with the value of 0 or \times whichever is the greater or the fewer ones, respectively; α , β , and ϵ are coefficients with values of 1, 0, and 0.1. In this way, a relevance score unit vector $\hat{\mathbf{R}}_{\text{cell}}$ corresponding to the curve \mathbf{Y}_{cell} can be obtained, where \mathbf{Y}_{cell} can be the

curves of voltage, current, capacity, and temperature of a given cell. The current DNNs can then be back-propagated layer by layer, turning a black-box into an explainable model.

All training process was based on python 3.7 and implemented in Keras (v2.3.1) with Tensorflow GPU backend (v2.1.0). Bayesian optimization with tree parzen estimator [31] technique has applied to all DNNs to optimize the hyper-parameters, such as the number of filters, kernel size, pooling type, and the suitable weight for the regularizer of layers. The process included 80 trials and each of which consisted of 80 epochs. All DNNs were trained by Amsgrad [32] optimizer with the default setting. We performed 5-fold cross-validation where 20% of the dataset was chosen as testing sets and 80% as training sets. During the training of the networks, spatial dropout [33] and early stopping [34] methods were adopted to prevent overfitting and to terminate the training. The specification of the current DNNs was detailed in Tables in Supplementary.

4.2. Data and codes

- The codes for prediction and the two experiments in supplementary are released in GitHub: https://github.com/acctouhou/Prediction_of_battery
- The entire pre-trained models and dataset of this work are available as follow: <https://drive.google.com/drive/folders/1Aq-wfoQ8ltDqziyHUcka7oUncQ7NgSP8?usp=sharing>

CRediT authorship contribution statement

Chia-Wei Hsu: Writing – original draft, Conceptualization, Methodology, Software, Validation, Visualization. **Rui Xiong:** Conceptualization, Validation. **Nan-Yow Chen:** Conceptualization, Methodology, Validation. **Ju Li:** Conceptualization, Methodology, Writing – review & editing, Project administration, Validation. **Nien-Ti Tsou:** Writing – original draft, Conceptualization, Project administration, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.apenergy.2021.118134>.

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