Remaining useful life and state of health prediction for lithium batteries based on differential thermal voltammetry and a deep learning model

Deep-learning model for battery SOH and RUL prediction

Feature extraction
- Degradation data
  - Voltage
  - Temperature

SG based DTV

Features Extraction
- PV1
- PV2
- PV3
- PV4
- Battery

Deep-learning model
- Model structure
  - Input layer
  - LSTM layers
  - Dropout layers
  - Output layer

Performance prediction
- Real SOH
- Simulated SOH
- RUL
- Maxwell

Error distribution

Highlights
- DTV captures phase transitions characterization in electrode materials
- Bayesian optimization can approach hyperparameters search of model
- Deep learning model can approach accurate estimation of battery SOH and RUL
- Deep learning model has excellent robustness with 40% missing data

Zhang et al., iScience 25, 105638 December 22, 2022 © 2022 The Authors. https://doi.org/10.1016/j.isci.2022.105638
It is crucial to estimate the health conditions of lithium-ion batteries (LIBs) due to the degradation of cathode and anode materials. In this paper, a fusion of deep learning model and feature analysis methods is employed to approach accurate estimation for state of health (SOH) and remaining useful life (RUL). The differential thermal voltammetry (DTV) signal analysis is executed to pre-process the datasets from Oxford University. A deep learning model is constructed with LSTM network as the core, combined with Bayesian optimization and dropout technique. This work shows that the deep learning model could approach the SOH and RUL early estimation with the mean absolute error of RUL maintained around 0.5%. It is potential that this deep learning model, combined with DTV signal analysis methods, could approach early prediction and estimation of battery SOH and RUL, contributing to the development of the next-generation high-energy-density and highly safety commercial batteries.
The construction process of the accurate battery mechanism model is relatively time-consuming and computationally expensive, which requires a professional knowledge background in electrochemistry.

Feature signal analysis methods, based on electrochemical analysis techniques and data processing methods, are expected to provide a signal characterization of battery degradation. There are some commonly used feature signal analysis methods, including electrochemical impedance spectroscopy, differential voltage analysis, and incremental capacity analysis. Hu et al. was concerned with machine learning-enabled battery SOH indication and prognosis. The advanced sparse Bayesian predictive modeling methodology is employed to capture the underlying correspondence between the capacity fade and sample entropy. Zhang et al. proposed a multi-objective decision method for data-driven-based estimation of battery states. However, the feature signal analysis methods require support of complex experiments. Besides, the accuracy is easily affected by uncontrolled noise in collected data under different external operating conditions.

The data-driven methods are exclusively dependent on historical experimental data to approach the SOH prediction and do not require much knowledge about battery internal mechanisms. Fei et al. proposed a comprehensive machine learning-based framework to achieve an accurate early-cycle prediction of battery lifetime. Ma et al. proposed a hybrid neural network with the false nearest neighbors methods. Patil et al. proposed a novel method for real-time RUL estimation of lithium ion batteries, integrating the classification and regression attributes of support vector-based machine learning techniques. Liu et al. combined the indirect health indicator and multiple Gaussian process model for RUL forecast to solve the capacity unmeasurable problem. Feng et al. reported an accurate and reproducible approach on how the data are processed with the simple code, exact fitting, computational availability, and reliability. Yang et al. proposed and constructed a hybrid neural network for battery monitoring and prognostics, combining the convolutional neural network and bidirectional long short-term memory network. Zhang et al. generated comprehensive dataset with 104 commercial batteries to establish a convolutional NN model for cycle life prediction. The data-driven methods have the highest implement ability, making their utilization more widespread. However, it is obvious that these methods have a bad interpretability and are highly dependent on the quality of the input data features. As for the battery degradation data, which is naturally timeseries, the long short-term memory (LSTM) model could be suitable for the estimation of battery SOH and RUL and therefore the LSTM network is utilized as the core in this paper.

The current approaches are either poorly interpretable or difficult to construct, so that none of them can be applied alone to meet the demands for highly accurate estimation and prediction of battery SOH and RUL at different scenarios. With the rapid increase in amount of available experiment data, the deep learning methods are emerging and suitable to deal with these high-dimensional data and further extract the main feature components. The deep learning model could approach battery degradation estimation and mine the internal correlations between battery operational characteristics and battery health conditions, not requiring complex knowledge of the internal mechanisms. However, the deep learning methods are also less interpretable and highly dependent on the input features. To compensate these drawbacks, the variable features that strongly correspond to battery degradation are required to be utilized for training and testing the model. That is, an effective solution is the utilization of multi-model fusion estimation methods to integrate the advantages of different approaches. The differential thermal voltammetry (DTV) method, as a kind of feature signal analysis method, could approach the link between macroscopic signal characteristics and microscopic battery degradation characteristics. Microscopically, the battery degradation could be characterized as the types and degree of internal phase transition in cathode and anode materials. Macroscopically, the external signal characteristics could be obtained and measured to observe the phase transition characteristics. That is, the change of battery surface temperature could correspond to the degree of phase transitions and the change of voltage could correspond to the type of phase transitions in electrode materials. On the one hand, the deep learning method can approach the accurate estimation of battery health conditions due to its excellent computing ability. On the other hand, the DTV method can mine the macroscopic signal characteristics for microscopic degradation characteristics, making deep learning methods interpretable. Therefore, with the utilization of DTV method, the deep learning model could have both the high implementation and the high interpretability.

In this paper, a fusion of deep learning method and feature signal analysis method is applied to approach the estimation and prediction of battery health conditions. The DTV signal analysis, which could
characterize the microscopic degradation characteristics, is executed to pre-process the datasets from Oxford University. With Savitzky-Golay (SG) filter method and Pearson correlation analysis, some variable features, which highly correspond to battery degradation process, are extracted and screened from DTV curves. Then a deep learning model is constructed with the LSTM network as the core. Besides, the Bayesian optimization and dropout technique are applied to optimize the hyperparameters and avoid the overfitting problems. This work shows that the deep learning model could approach the long-term SOH and RUL early estimation and prediction with the input features extracted from DTV curves. It is potential that this deep learning model, combined with DTV signal analysis methods, could approach the early prediction and estimation of SOH and RUL, contributing to the development of the next-generation high-energy-density and highly safety commercial batteries.

Data pre-processing
In this section, the DTV signal analysis is carried out based on the datasets from Oxford University, and the highly correlated features are extracted and screened with SG method and Pearson correlation analysis.

Battery degradation experimental data analysis
In this paper, the battery degradation dataset from Oxford University is utilized to train and validate the deep learning model. The experimental battery electrodes include graphite anodes, lithium cobalt oxide, and lithium nickel cobalt oxide cathodes (LCO/NCO). There are 8 groups of 740 mAh pouch batteries (labeled from battery #1 to #8). However, the battery capacity of battery #2 and #5 drop sharply and the surface temperatures change significantly. The battery #6 does not reach below the end of life. Consequently, five of 8 battery groups, battery #1, #3, #4, #7, and #8 are selected and utilized for model training and validating. Table 1 shows the technical specifications and battery test procedures of the five batteries. Figure 1A shows the completed degradation cycle test for voltage, current, and temperature. Figure 1B shows the capacity degradation curves of the five batteries. It could be seen that the downward trend of battery #4 is significantly faster than those of the other four groups of batteries. Therefore, to obtain reliable prediction of battery health conditions, the robust features should be extracted and screened.

Figure 2 shows the evolution patterns of the open-circuit voltage and surface temperature of battery #1 during the degradation process. It could be seen that with the battery degradation, both the open-circuit voltage and surface temperature deliver an obvious trend of change. That is, the external signal characteristics could reflect the internal battery degradation characteristics. However, considering the complexity of the real vehicle discharge conditions and the relatively uniform charging conditions, the charge cycle data are utilized for SOH prediction in this paper.

Differential thermal voltammetry curve analysis and filter
In this subsection, the DTV signal analysis method is described in detail and executed combined with SG method to obtain the smooth DTV curves.

The DTV method, proposed by Wu et al. in 2015, could help to extract feature variables that could reflect microscopic battery degradation characteristics. The DTV could be calculated as follows:

$$ DTV = \frac{dT}{dt} \div \frac{dV}{dt} = \frac{dT}{dV} $$  \hspace{1cm} (Equation 1)
where $T$ represents the battery surface temperature, $V$ the terminal voltage of battery, and $t$ the sample time. Considering the entropic changes coupling with battery system, the DTV technique could be utilized in this work to evaluate the change patterns of external signal characteristics during degradation cycles. That is, the utilization of DTV technology could bridge macroscopic signal characteristics and microscopic phase transition characteristic. The DTV method tracks battery degradation by tracking phase transitions and the entropic heat generated in the electrodes, providing information on battery degradation on a shorter timescale and in a simpler experimental environment, similar to that of slow cyclic voltammetry and incremental capacity analysis. As shown in Figures 3C and 3D, there are different phase combinations during the battery degradation process in electrode materials. The positive and negative electrodes are in different phase combinations throughout the battery degradation process. And the transition between these phases involves a positive and negative phase transition, which could be accompanied by an entropic abrupt change in the form of an inflection point on the DTV curves. That is, the peaks and valleys of DTV curves could reflect the current position of the phase transitions in the cathode and anode. Specifically, the peak position describes the peak potential at which the (de)intercalation stages are occurring. The shift in the peak position through degradation can describe both the impedance rise of the cell and stoichiometric drift. Peak height indicates the point of maximum rate of heat generation for those phases. Peak width describes the potential window of the combined phases in the two electrodes. Peak area gives information on the heat generated during the (de)intercalation stages. Besides, phase transitions require energy to drive them, leading to changes in temperature. That is, the change of battery surface temperature indicates the degree of phase transitions, and the change of terminal voltage indicates the type of phase transitions. Therefore, the DTV methods can bridge the macroscopic signaling characteristics and microscopic degradation characteristics through the articulation of phase transitions characteristics based on the datasets of temperature and terminal voltage, which are relatively easy to measure.

Figure 3A demonstrates the comparison of temperature curves before and after smoothing by SG method. Figure 3B shows the comparison of initial DTV curve, DTV curve after temperature smooth, and DTV curve smoothed by SG method. It could be seen that the DTV curves have smaller fluctuations after smoothed by SG method.
Feature extraction of battery degradation and correlation analysis

In this subsection, the highly correlated features are extracted and screened with Pearson correlation analysis method. As shown in Figure 3D, the DTV curves during the degradation process have one valley and two peaks (labeled peak1 and peak2). It could be seen that the evaluation patterns of coordinates of wave peaks and valley demonstrate a certain directionality. The horizontal coordinates of these peaks change in an increasing direction. The horizontal coordinate of the valley changes in a decreasing direction. Based on the mechanism of DTV methods analyzed in section differential thermal voltammetry curve analysis and filter, here 6 features are extracted from peaks and valley of DTV curves. As shown in Figure 4A, the coordinates of the peak1, valley, and the peak2 are extracted as the variable features \([F_1, F_2]\), \([F_3, F_4]\), and \([F_5, F_6]\).

The specific mathematical description of the peaks and valley could be expressed as follows:

\[
\begin{align*}
V_{\text{peak}} &= V_{|\Delta V| = 0, \text{ and } f(V) \geq f(V), V \in (V_{i-1}, V_{i+1})} \\
DTV_{\text{peak}} &= f(V_{\text{peak}}) \\
\end{align*}
\] (Equation 3)

\[
\begin{align*}
V_{\text{valley}} &= V_{|\Delta V| = 0, \text{ and } f(V) \geq f(V), V \in (V_{i-1}, V_{i+1})} \\
DTV_{\text{valley}} &= f(V_{\text{valley}}) \\
\end{align*}
\] (Equation 4)

where \(f(\cdot)\) represents a mapping function between voltage and DTV, and \(V_{i-1}\) and \(V_{i+1}\) the voltages of the previous sampling time point and the later sampling time point, respectively.

Figures 4B and 4C demonstrate the changes of these six features of battery #1 with the cycle number. It could be seen that the data of F2, F4, and F5 show more stable and smooth characteristics. The fitting of the data points could be closer to a straight line. The data of F4 and F5 demonstrate a positive correlation and the data of F2 demonstrate a negative correlation. Then the Pearson correlation analysis is executed to further analyze the correlation between the six feature variables and the capacity fade. The formula of Pearson correlation analysis could be described as follows:

\[
\begin{align*}
r_{xy} &= \frac{\sum_{i=1}^{n} (x_i - \bar{x})(y_i - \bar{y})}{\sqrt{\sum_{i=1}^{n} (x_i - \bar{x})^2 \sum_{i=1}^{n} (y_i - \bar{y})^2}} \\
\end{align*}
\] (Equation 5)

where \(n\) represents the number of sample series, \(x\) and \(y\) respectively represent a variable, and \(\bar{x}\) and \(\bar{y}\) respectively denote mean values of \(x\) and \(y\). Figures 4D and 4E demonstrate the correlation matrix of battery #1 and battery #8, respectively. The flatter the oval, the higher the correlation between the two variables. It could be seen that the F2, F4, and F5 show a strong correlation with battery capacity degradation, whose absolute values of correlation coefficients are all close to 1. Therefore, the three features, F2, F4, and F5, are utilized as the input features to train the deep learning model for battery degradation prediction.

Figure 2. Evolution of the open-circuit voltages and surface temperatures of battery #1 throughout the degradation process
(A) Surface temperature.
(B) Open-circuit voltage.
Methodologies

In this section, the framework and process of the proposed deep learning model for battery degradation prediction is described in detail, including the LSTM network as the core, the model training process, and some error analysis methods.

Framework of the proposed RUL & SOH estimation model

Figure 5 demonstrates the specific framework of the LSTM-based battery SOH and RUL prediction model. The framework is divided into three parts, including feature extraction, model construction & training, error analysis, respectively.

In the first part, the feature extraction is executed. Three highly correlated feature variables are extracted and screened from DTV curves to train and test the data-driven model as described in detail in section data pre-processing. In the second part, a data-driven model with LSTM network as the core is constructed. The three-dimensional timeseries tensor obtained in the first part is applied as input to train the model. The Bayesian optimization method is utilized to approach hyperparameters search. The RMSprop technique is applied to improve the convergence speed during training. The dropout technique is introduced to overcome the overfitting. Finally, four error analysis methods are utilized to quantitatively analyze the prediction results of model, including absolute error, mean absolute error, root-mean-square error analysis, and box-plot analysis.

Input and output data structure

The time series is constructed to follow the feature extraction, that is, the timeseries data are constructed based on the three features F2, F4, and F5 extracted in section data pre-processing and the capacity.
input of the deep learning neural network is a three-dimensional timeseries tensor. Three feature variables are utilized to construct the input dataset. Firstly, each of the charge and discharge cycle extracts a one-dimensional vector containing four variables, including three input features from DTV curves and one label from SOH data. The combination of vectors of different cycles could form a two-dimensional matrix. Then according to the sequence length, two-dimensional data fragments are extracted from the above two-dimensional matrix. Finally, according to the batch size, the two-dimensional data fragments above are combined to build a three-dimensional tensor as the final input dataset.

Long short-term memory neural network

The cell states of LSTMs can selectively remember or forget information. There exist three types of gates that can work for the cell state, including forget gate, input gate, and output gate. Forget gate could remove the information which is no longer important and needed. Input gate could receive the addition of information. Output gate is responsible for selecting useful information and generating the output. The LSTM model can be built by creating three layers with the help of three gates.
The forward transfer process of the LSTM unit at time $t$ can be described as follows:

\[
\begin{align*}
f_k &= \sigma(W_f \cdot [x_k, h_{k-1}] + b_f) \\
\bar{G}_k &= \tanh(W_c \cdot [x_k, h_{k-1}] + b_c) \\
G_k &= f_k \cdot G_{k-1} + i_k \cdot \bar{G}_k \\
h_k &= o_k \cdot \tanh(G_k)
\end{align*}
\]

(Equation 6)

(Equation 7)

(Equation 8)

(Equation 9)

(Equation 10)

(Equation 11)

where $i_k$, $f_k$, and $o_k$ represent the activation vectors of the input gate, forget gate, and output gate, respectively. $\sigma$ is the sigmoid activation function. $G_k$ represents the long-term memory stored in a cell. $h_k$ can store short-term memory. $\bar{G}_k$ is the candidate state. $W_f$, $W_i$, $W_o$, and $W_c$ are related to the weight of forget gate, input gate, output gate, and unit gate, respectively. $b$ is the deviation value.

**Model training**

During the model training process, the Bayesian optimization, the RMSprop algorithm, and the dropout techniques are utilized to improve the model performances.

The Bayesian optimization method is applied for hyperparameter optimization during model training process. The Bayesian optimization could be considered as the most advanced optimization framework. The hyperparameters, which generally could not be directly optimized using conventional optimization tools, that is, the pre-set parameters before training the deep learning model rather than the parameter obtained through training. The hyperparameters can have a great impact on the deep learning model performances. However, the value of hyperparameters cannot be utilized for different types of batteries to meet the increasing demands of high prediction accuracy. The Bayesian optimization methods could automatically adjust hyperparameters to obtain the best performance of neural network. In this paper, the hyperparameters that could be optimized include the learning rate and the rates of three dropout layers. The calculation could be computed as follows:

\[
d^* = \arg\min J(d), D \in (a, b), (d \in D)
\]

(Equation 12)
where $d$ represents the hyperparameter value, $d^*$ the optimal value, and $(a, b)$ the interval of optimization.

The RMSprop algorithm has been empirically proven to be an effective and practical algorithm for deep learning neural network optimization. It works very well for RNNs, and is one of the frequently adopted optimization methods nowadays, which can further improve convergence speed and convergence character.

The dropout technique can be applied to overcome the problems of overfitting during the training process. Specifically, the dropout layer could randomly drop neurons and their connections, that is, the dropout technique can reduce the interaction between hidden layer nodes by randomly dropping some of the hidden layer nodes with a certain probability. Noting that the dropout technique has a deterministic index of what percentage of all hidden nodes could be dropped but the specific nodes that are discarded are chosen randomly.

**Error analysis**

In this paper, four evaluation indicators are utilized to carry out the error analysis, including absolute error, mean absolute error (MAE), root-mean-square error (RMSE), and box-plot analysis, respectively.

The absolute error means the difference between an observed value of a quantity and the true value. The calculation could be described as follows:

$$
\Delta x = x_0 - x \quad \text{(Equation 13)}
$$

where $x_0$ represents the measured or inferred value of a quantity and $x$ the actual value.

The MAE is the average difference between the observations/true values and model output or predictions. The MAE could be expressed as follows:

$$
\text{MAE} = \frac{1}{N} \sum_{i=1}^{N} |y_{i,\text{pred}} - y_{i,\text{true}}| \quad \text{(Equation 14)}
$$

where $y_{i,\text{pred}}$ represents the predicted values, $y_{i,\text{true}}$ the observations, and $N$ the total number of samples.

The RMSE is the sum and then average of the squares of the difference between predicted values and true values. The RMSE could be calculated as follows:

$$
\text{RMSE} = \sqrt{\frac{1}{N} \sum_{i=1}^{N} (Y_i - f(x_i))^2} \quad \text{(Equation 15)}
$$

where RMSE is the corresponding value for analysis of RMSE. The $Y_i$ and $f(x_i)$ are the actual measured values and predicted values, respectively.

Box-plot analysis consists of five parts, the minimum number, the maximum number, the lower quartile, the median number, and the upper quartile. Boxplots could effectively help us visually identify the characteristics of the data.

**RESULTS AND DISCUSSION**

In this section, the constructed model is trained and validated based on the datasets from Oxford University. Besides, the robustness of the model is verified to prove the early prediction ability of the proposed model.

**Effect of hyperparameter optimization and number of LSTM layers**

In this subsection, the effect of Bayesian optimization method and the number of LSTM layers is investigated. The experimental batteries consist of graphite anodes, lithium cobalt oxide, and lithium nickel cobalt oxide cathodes (LCO/NCO). The batteries are cycled through an 8-channel Big MPG 205 battery tester and housed in an MKS3 hot chamber at 40°C with constant current charge and discharge. Based on the datasets of these batteries, a total of four prediction results are shown, including the Case #1, #2, #3, and #4, respectively. Table 2 demonstrates the specific experimental structure and setting of the four cases.
Table 2. Experimental control group – Bayesian optimization and LSTM layers

| Items                                | Case #1                             | Case #2                             | Case #3                             | Case #4                             |
|--------------------------------------|-------------------------------------|-------------------------------------|-------------------------------------|-------------------------------------|
| Bayesian hyperparameter optimization | ✓                                   | ×                                   | ✓                                   | ✓                                   |
| Number of dropout layers             | 2                                   | 2                                   | 3                                   | 1                                   |
| Model construction & activation function | ![Diagram 1](image1) | ![Diagram 2](image2) | ![Diagram 3](image3) | ![Diagram 4](image4) |
| Units of layers                      | [56, -, 64, -, 1]                   | [64, -, 64, -, 1]                   | [92, -, 38, -, 114, - 1]            | [18, -, 1]                         |
| Rate of dropout layer 1             | 0.174                               | 0.2                                 | 0.376                               | 0.264                               |
| Rate of dropout layer 2             | 0.249                               | 0.2                                 | 0.417                               | –                                   |
| Rate of dropout layer 3             | –                                   | –                                   | 0.531                               | –                                   |
| Learning rate                       | 0.00352                             | 0.001                               | 0.00425                             | 0.00879                             |
| Optimizer                            | RMSprop                             | RMSprop                             | RMSprop                             | RMSprop                             |
| Loss                                 | Mean squared error                  | Mean squared error                  | Mean squared error                  | Mean squared error                  |
| Sequence length                      | 2                                   | 2                                   | 2                                   | 2                                   |
| Batch Size                           | 2                                   | 2                                   | 2                                   | 2                                   |
| Epoch size                           | 50                                  | 50                                  | 50                                  | 50                                  |
Case #1, #3, and #4 all apply the Bayesian optimization methods but Case #2 does not. Case #1 and Case #2 utilize 2 layers of LSTM network. Case #3 utilizes three layers of LSTM network and Case #4 utilizes one layer of LSTM network.

Figure 6 shows the estimation results and error analysis of four cases. The black line represents the real SOH while the hollow circles represent the estimated SOH. The real RUL and estimated RUL of the battery are illustrated in each subgraph. Besides, the division proportion of the dataset is marked obviously.

In Case #1, the estimated RUL is 3400 cycles and the real RUL is 3300 cycles. In Case #2, the estimated RUL is 2900 cycles and the real RUL is 3300 cycles. In Case #3, the estimated RUL is 3200 cycles and the real RUL is 3300 cycles. In Case #4, the estimated RUL is 3200 cycles and the real RUL is 3300 cycles. Figures 6A and 6B demonstrate the estimation results of Case #1 and #2. It could be seen that the prediction results of Case #1 show a higher accuracy with the MAE of 0.44%, which could be proven in Figures 6E and 6F as well. That is, the utilization of Bayesian optimization method could improve the accuracy of model. Compared with the estimation results of Case #1, the prediction results of Case #3 and #4, which have a different setting on
number of LSTM layers, show a lower accuracy with the MAE of 1.11% and 0.64%, respectively. Figures 6E and 6F could demonstrate that model with the experimental setting of Case #1 has the highest accuracy. Therefore, in the following experiments, the number of LSTM layers is selected to be equal to 2. The results show that our model could achieve accurate SOH estimation and RUL prediction.

**Estimation results in different batteries**

In this subsection, the constructed and trained model is validated based on the datasets from different batteries. As shown in Figure 7, the validation and error analysis are executed on the datasets from battery #3, battery #4, battery #7, and battery #8. The entire prediction errors are distributed around 0.5%. As shown in Figure 7A, the estimated RUL is 3600 cycles and the real RUL is 3800 cycles. Figure 7B shows that the real RUL is 3000 cycles and the estimated RUL is 2900 cycles. Figure 7C shows that the real RUL is 5300 cycles and the estimated RUL is 5200 cycles. Figure 7D shows that the real RUL is 3900 cycles and the estimated RUL is 3800 cycles. It could be seen that the estimated results do not exactly match the real results but are within a
tolerable level of accuracy. The reason for the errors may be that the original dataset is in a large 100-cycles increment, which could result in some information missing. Besides, the estimated RUL tends to be lower than the real RUL, which could benefit the safety management in practical applications.

Figures 7E and 7F show the errors of the model prediction results based on the datasets from four batteries. The minimum MAE could reach 0.3% and the maximum MAE could reach 0.61%, which further proving the accuracy and stability of our model. Overall, it is validated that the proposed model can approach the accurate estimation and prediction of battery health conditions.

Verification of the robustness of the proposed model
The robustness is the ability of the system to survive in unusual and dangerous situations. In practical problems, the disturbances in system characteristics or parameters are often unavoidable. The robustness of a system is the key to its real practical application. In this subsection, the validation of the robustness and reliability of the model is executed based on the datasets from different batteries. As shown in Figure 8, the division proportion of the datasets is set with 40% missing data, 12% training data, and 48% testing data.
data. Figure 8A shows that the real RUL is 1200 cycles and the estimated RUL is 1100 cycles. Figure 8B shows that the real RUL is 1400 cycles and the estimated RUL is 1300 cycles. Figure 8C shows that the real RUL is 2700 cycles and the estimated RUL is 2100 cycles. Figure 8D shows that the real RUL is 1500 cycles and the estimated RUL is 1700 cycles. Besides, it could be seen that the estimated SOH could match the real SOH well for four prediction results even with 40% missing data.

Figures 8E and 8F demonstrate the box-plot analysis and error analysis for the four prediction results. The model could produce a prediction MAE of around 0.5% with 40% missing data, which is not significantly different from the result accuracy without data loss. The minimum MAE could reach 0.38% and the maximum MAE could reach 0.63%. It could be seen that the missing data do not interfere much with the accuracy of the model. Therefore, the deep learning model with two layers of LSTM network has excellent robustness and stability with the help of Bayesian optimization methods.

Conclusions

In this paper, a data-driven model with LSTM network as the core, combined with feature signal analysis methods, is constructed and trained to approach the estimation and prediction of battery SOH and RUL. The DTV signal analysis is executed based on the battery voltage and temperature information from Oxford University. Then three feature variables are extracted and screened as the input of the following model with the SG filter method and Pearson correlation analysis. Subsequently, a data-driven model is constructed with the dropout techniques and Bayesian optimization introduced to improve the model performances. The DTV signal analysis methods, which can bridge microscopic phase transition in electrode materials and macroscopic signal characteristics, can solve the problems that the data-driven model tends to be less interpretable and highly dependent on the quality of the input data features.

The effect of the Bayesian optimization for the hyperparameters and the number of LSTM layers are investigated firstly. The results show that the model with two LSTM layers could have a highest accuracy with an MAE of 0.44% supported by Bayesian optimization. Then the model is validated based on the datasets from four different batteries. Finally, the robustness and reliability of our model is verified. The MAE can be maintained around 0.5% with 40% missing data. The results show that the data-driven model can approach the accurate and stable prediction of the battery SOH and RUL.

Overall, the DTV methods can characterize the microscopic phase transitions in cathode and anode materials to capture the degradation characterization based on the macroscopic signaling characteristics. Three highly correlated feature variables are extracted and screened with the DTV signal analysis methods and SG filter methods. The Bayesian optimization method is utilized to approach the hyperparameters search. The data-driven model constructed with LSTM network as the core can approach the early and robust prediction of battery health conditions. It’s potential that our model can contribute to highly accurate estimation and safety management of battery full life cycle with the powerful computing ability on the cloud platform.

STAR METHODS

Detailed methods are provided in the online version of this paper and include the following:

- **KEY RESOURCES TABLE**
- **RESOURCE AVAILABILITY**
  - Lead contact
  - Materials availability
  - Data and code availability
- **METHOD DETAILS**
  - Differential thermal voltammetry signal analysis
  - Bayesian optimization
  - Dropout technique
  - RMSprop algorithm

ACKNOWLEDGMENTS

This work was financially supported by the National Natural Science Foundation of China (No. 52102470).
AUTHOR CONTRIBUTIONS

Conceived and designed the experiments and analyzed data: L.Z., Z.Z., and X.L.; Performed most of experiments: L.Z., W.W., and H.Y.; Writing – Original Draft: L.Z.; Writing – Review & Editing: X.Y., F.L., and S.L.; Conceptualization, Methodology: S.Y., Z.Z., and X.L.; All authors contributed to and approved the paper.

DECLARATION OF INTERESTS

The authors declare no competing interests.

Received: August 18, 2022
Revised: September 13, 2022
Accepted: November 17, 2022
Published: December 22, 2022

REFERENCES

1. Zhang, L.-S., Gao, X.-L., Liu, X.-H., Zhang, Z.-J., Cao, R., Cheng, H.-C., Wang, M.-Y., Yan, X.-Y., and Yang, S.-C. (2022). CHAIN: unlocking informatics-aided design of Li metal anode from materials to applications. Rare Met. 41, 1477–1489. https://doi.org/10.1007/s12598-021-01925-8.

2. Lu, Q., Jie, Y., Meng, X., Omar, A., Mikhailova, D., Cao, R., Jiao, S., Lu, Y., and Xu, Y. (2021). Carbon materials for stable Li metal anodes: challenges, solutions, and outlook. Carbon Energy 3, 957–975. https://doi.org/10.1002/cey2.147.

3. Chen, X.R., Zhao, B.C., Yan, C., and Zhang, Q. (2021). Review on Li deposition in working batteries: from nucleation to early growth. Adv. Mater. 33, e20004128. https://doi.org/10.1002/adma.2020004128.

4. Cao, R., Cheng, H., Jia, X., Gao, X., Zhang, Z., Wang, M., Li, S., Zhang, C., Ma, B., Liu, X., and Yang, S. (2022). Non-invasive characteristic curve analysis of lithium-ion batteries enabling degradation analysis and data-driven model construction: a review. Automot. Innov. 5, 146–163. https://doi.org/10.1007/s42154-022-00181-5.

5. Makwarumba, C.P., Tang, M., Peng, Y., Lu, S., Zheng, L., Zhao, Z., and Zhen, A.G. (2022). Assessment of recycling methods and processes for lithium-ion batteries. iScience 25, 104321. https://doi.org/10.1016/j.isci.2022.104321.

6. Liu, Y., Zhang, R., Wang, J., and Wang, Y. (2021). Current and future lithium-ion battery manufacturing. iScience 24, 102332. https://doi.org/10.1016/j.isci.2021.102332.

7. Meng, X., Xu, Y., Cao, H., Lin, X., Ning, P., Zhang, Y., Garcia, Y.G., and Sun, Z. (2020). Internal failure of anode materials for lithium batteries — a critical review. Green Energy Environ. 5, 22–36. https://doi.org/10.1007/s42154-020-00166-w.

8. Elattar, H.M., Elminir, H.K., and Riad, A.M. (2016). Prognostics: a literature review. Complex Intell. Syst. 2, 125–154. https://doi.org/10.1007/s40747-016-0019-3.

9. Xiong, R., Ma, S., Li, H., Sun, F., and Li, J. (2020). Toward a safer battery management system: a critical review on diagnosis and prognosis of battery short circuit. iScience 23, 105638, December 22, 2022. https://doi.org/10.1016/j.isci.2020.105638.

10. Yang, S., He, R., Zhang, Z., Cao, Y., Gao, X., and Liu, X. (2020). CHAIN: cyber hierarchy and interactional network enabling digital solution for battery full-lifespan management. Matter 3, 27–41. https://doi.org/10.1016/j.matt.2020.04.015.

11. Tao, H., Lian, C., and Liu, H. (2020). Multiscale modeling of electrolytes in porous electrode from equilibrium structure to non-equilibrium transport. Green Energy Environ. 5, 303–321. https://doi.org/10.1007/j.2020.06.020.

12. Yang, Z., Patil, D., and Fahimi, B. (2018). Online estimation of capacity fade and power fade of lithium-ion batteries based on input-output response technique. IEEE Trans. Electrific. 4, 147–156. https://doi.org/10.1109/TTE.2017.2758301.

13. Li, Y., Liu, K., Foley, A.M., Zülke, A., Bercicar, M., Narini-Masyr, E., Van Mierlo, J., and Hosten, H.E. (2019). Data-driven health estimation and lifetime prediction of lithium-ion batteries: a review. Renew. Sustain. Energy Rev. 113, 109254. https://doi.org/10.1016/j.rser.2019.109254.

14. Lin, C., Kong, W., Tian, Y., Wang, W., and Zhao, M. (2022). Heating lithium-ion batteries at low temperatures for onboard applications: recent progress, challenges and prospects. Automot. Innov. 5, 3–17. https://doi.org/10.1007/s42154-022-00166-w.

15. Feng, Y., Xue, C., Han, Q.L., Han, F., and Du, J. (2020). Robust estimation for state-of-charge and state-of-health of lithium-ion batteries using integral-type terminal sliding-mode observers. IEEE Trans. Ind. Electron. 67, 4013–4023. https://doi.org/10.1109/TIE.2019.2916389.

16. Meng, J., Cai, L., Stroe, D.I., Ma, J., Luo, G., and Teodorescu, R. (2020). An optimized ensemble learning framework for lithium-ion Battery State of Health estimation in energy storage system. Energies 206, 118140. https://doi.org/10.1016/j.ener2020.118140.

17. Keil, P., Schuster, S.F., Wilhelm, J., Travi, J., Hauser, A., Karl, R.C., and Jossen, A. (2016). Calendar aging of lithium-ion batteries. J. Electrochem. Soc. 163, A1872–A1880. https://doi.org/10.1149/2.041160jes.

18. Cai, Y., Yang, L., Deng, Z., Zhao, X., and Deng, H. (2018). Online identification of lithium-ion battery state-of-health based on fast wavelet transform and cross D-Markov machine. Energy 147, 621–635. https://doi.org/10.1016/j.energy.2018.01.001.

19. Zhou, C.C., Su, Z., Gao, X.L., Cao, R., Yang, S.C., and Liu, X.H. (2022). Ultra-high-energy lithium-ion batteries enabled by aligned structured thick electrode design. Rare Met. 41, 14–20. https://doi.org/10.1007/s12598-021-01785-2.

20. Jokar, A., Rajabloo, B., Désilets, M., and Lacroix, M. (2016). Review of simplified Pseudo-two-Dimensional models of lithium-ion batteries. J. Power Sources 327, 44–55. https://doi.org/10.1016/j.jpowsour.2016.07.036.

21. Li, J., Adewuyi, K., Lofti, N., Landers, R.G., and Park, J. (2018). A single particle model with chemical/mechanical degradation physics for lithium ion battery State of Health (SOH) estimation. Appl. Energy 212, 1178–1190. https://doi.org/10.1016/j.apenergy.2018.01.011.

22. Baghdadi, I., Briat, O., Delétage, J.Y., Gyan, P., and Vinassa, J.M. (2016). Lithium battery aging model based on Dain’s degradation approach. J. Power Sources 325, 273–285. https://doi.org/10.1016/j.jpowsour.2016.06.036.

23. Wang, J., Purewal, J., Liu, P., Hicks-Garnier, J., Soukazian, S., Sherman, E., Sorenson, A., Vu, L., Tataria, H., and Verbrugge, M.W. (2014). Lithium battery SOH estimation. Appl. Energy Rev. 23, 102332. https://doi.org/10.1016/j.apenergy.2016.07.006.

24. Gopaluni, R.B., and Braatz, R.D. (2013). State of charge estimation in Li-ion batteries using an isothermal pseudo two-dimensional model. IFAC Proc. Vol. 46, 135–140. https://doi.org/10.3182/20131218-3-IN-2045.00163.
25. Ecker, M., Gerschler, J.B., Vogel, J., Käbitz, S., Hurt, F., Dechent, P., and Sauer, D.U. (2012). Development of a lifetime prediction model for lithium-ion batteries based on extended accelerated aging test data. J. Power Sources 215, 248–257. https://doi.org/10.1016/j.jpowsour.2012.05.012.

26. Tian, J., Xu, R., Wang, Y., and Chen, Z. (2021). Capacity attenuation mechanism modeling and health assessment of lithium-ion batteries. Energy 227, 119682. https://doi.org/10.1016/j.energy.2020.119682.

27. Liu, X., Zhang, L., Yu, H., Wang, J., Li, J., Yang, K., Zhao, Y., Wang, H., Wu, B., Brandon, N.P., et al. (2022). Bridging multiscale characterization technologies and digital modeling to evaluate lithium battery full lifecycle. Adv. Energy Mater. 12, 2200889. https://doi.org/10.1002/aenm.202200889.

28. Hu, X., Jiang, J., Cao, D., and Egardt, B. (2013). Battery health prognosis for electric vehicles using sample entropy and sparse Bayesian predictive modeling. IEEE Trans. Ind. Electron. 63, 1–2656. https://doi.org/10.1109/TIE.2015.2461523.

29. Zhang, S., Guo, X., and Zhang, X. (2020). Multi-objective decision analysis for data-driven based estimation of battery states: a case study of remaining useful life estimation. Int. J. Hydrogen Energy 45, 14156–14173. https://doi.org/10.1016/j.ijhydene.2020.03.100.

30. Wu, L., Fu, X., and Guan, Y. (2016). Review of the remaining useful life prognostics of vehicle lithium-ion batteries using data-driven methodologies. Appl. Sci. 6, 166. https://doi.org/10.3390/app6060166.

31. Zhang, Y., Xiong, R., He, H., and Pecht, M.G. (2019). Lithium-ion battery remaining useful life prediction with box-cox transformation and Monte Carlo simulation. IEEE Trans. Ind. Electron. 66, 1585–1597. https://doi.org/10.1109/TIE.2018.2838918.

32. Ding, W.L., Lu, Y., Peng, X.L., Dong, H., Chi, W.J., Yuan, X., Sun, Z.Z., and He, H. (2021). Accelerating evaluation of the mobility of ionic liquid-modulated PEDOT flexible electronics using machine learning. J. Mater. Chem. 9, 25547–25557. https://doi.org/10.1039/d1ta08013c.

33. Huang, H., Meng, J., Wang, Y., Cai, L., Peng, J., Wu, J., Xiao, Q., Liu, T., and Teodorescu, R. (2022). An enhanced data-driven model for lithium-ion battery state-of-health estimation with optimized features and prior knowledge. Automot. Innov. 5, 134–145. https://doi.org/10.1007/s42154-022-00175-3.

34. Fei, Z., Yang, F., Tsui, K.L., Li, L., and Zhang, Z. (2021). Early prediction of battery lifetime via a machine learning based framework. Energy 225, 120205. https://doi.org/10.1016/j.energy.2021.120205.

35. Ma, G., Zhang, Y., Cheng, C., Zhou, B., Hu, P., and Yuan, Y. (2019). Remaining useful life prediction of lithium-ion batteries based on false nearest neighbors and a hybrid neural network. Appl. Energy 253, 113626. https://doi.org/10.1016/j.apenergy.2019.113626.

36. Patil, M.A., Tagade, P., Harirahan, K.S., Kolake, S.M., Song, T., Yeo, T., and Doo, S. (2018). A novel multistage Support Vector Machine based approach for Li ion battery remaining useful life estimation. Appl. Energy 159, 285–297. https://doi.org/10.1016/j.apenergy.2015.08.119.

37. Liu, J., and Chen, Z. (2019). Remaining useful life prediction of lithium-ion batteries based on health indicator and Gaussian process regression model. IEEE Access 7, 39474–39484. https://doi.org/10.1109/ACCESS.2019.2905740.

38. Feng, X., Merla, Y., Weng, C., Ouyang, M., He, X., Liaw, B.Y., Santhananokalan, S., Li, X., Liu, P., Lu, L., et al. (2020). A reliable approach of differentiating discrete sampled-data for battery diagnosis. eTransportation 3, 100051. https://doi.org/10.1016/j.etran.2020.100051.

39. Yang, H., Wang, P., An, Y., Shi, C., Sun, X., Wang, K., Zhang, X., Wei, T., and Ma, Y. (2020). Remaining useful life prediction based on denoising technique and deep neural network for lithium-ion capacitors. eTransportation 5, 100078. https://doi.org/10.1016/j.etran.2020.100078.

40. Su, L., Wu, M., Li, Z., and Zhang, J. (2021). Cycle life prediction of lithium-ion batteries based on data-driven methods. eTransportation 10, 100137. https://doi.org/10.1016/j.etran.2021.100137.

41. Wu, B., Yufit, V., Merla, Y., Martinez-Botas, R.F., Brandon, N.P., and Offer, G.J. (2015). Differential thermal voltammetry for tracking of degradation in lithium-ion batteries. J. Power Sources 273, 495–501. https://doi.org/10.1016/j.jpowsour.2014.09.127.

42. Birkl, C. (2017). Oxford Battery Degradation Dataset 1. VO - RT - Aggregated Database. OP -.

43. Birkl, C. (2017). Diagnosis and Prognosis of Degradation in Lithium-Ion Batteries. VO - RT - Thesis. OP -.
**STAR METHODS**

**KEY RESOURCES TABLE**

| REAGENT or RESOURCE | SOURCE | IDENTIFIER |
|---------------------|--------|------------|
| **Software and algorithms** | TensorFlow | https://tensorflow.google.cn/ |
| TensorFlow 2.3 | TensorFlow | https://tensorflow.google.cn/ |
| Python 3.8 | Python | https://www.python.org/ |

**RESOURCE AVAILABILITY**

**Lead contact**
Further information and requests for resources and reagents should be directed to and will be fulfilled by the lead contact, Xinhua Liu (liuxinhua19@buaa.edu.cn).

**Materials availability**
This study did not generate new materials.

**Data and code availability**
No additional data was used. This paper does not report original code. Any additional information for re-analyzing this work is available from the lead contact upon request.

**METHOD DETAILS**

**Differential thermal voltammetry signal analysis**
The DTV method is an important tool for tracking battery health conditions, which could be utilized to evaluate the patterns of battery degradation process. It can help to extract feature variables that reflect microscopic battery degradation characteristics. The battery degradation is a complex physicochemical process. Therefore, the analysis of internal mechanism and measurement of parameters could be challenging. The change of entropy is a function of temperature, and the DTV method could provide obvious information related to entropy. Some feature variables, such as positions and heights of peak and valleys of the curve, are directly concerned with the impedance increment and nonuniform of electrode performance during battery degradation process, reflecting the phase transition characteristic. The phase transition characteristic is closely related to the battery degradation, thus providing close links between DTV features and battery degradation. The parameters of DTV methods could be calculated by differentiate the temperature of the battery surface to the terminal voltage during charging, described as follows.

\[
\text{DTV} = \frac{dT}{dt} = \frac{dT}{dV} \quad \text{(Equation 16)}
\]

Where \( T \) represents the battery surface temperature, \( V \) the battery terminal voltage. That is, we could obtain DTV only by obtaining temperature and voltage data from the battery during charging or discharging.

**Bayesian optimization**
The Bayesian optimization method is utilized for hyperparameters search of model. As for deep-learning neural network, the hyperparameters are pre-set parameters rather than the parameter obtained through training. The hyperparameters have a great effect on the performance of neural networks. With the improvement of health prediction accuracy, the value of hyperparameters cannot be applied to different types of batteries. Reasonable selection of hyperparameters can optimize the results of network calculation. The hyperparameter value is automatically adjusted by Bayesian optimization algorithm. The optimal hyperparameter value could be computed as follows:

\[
d^* = \text{argmin}_{d} J(d), \quad D \in (a, b), \quad (d \in D) \quad \text{(Equation 17)}
\]

Where \( d \) represents the hyperparameter value, \( d^* \) the optimal value and \((a, b)\) the interval of optimization.
**Dropout technique**

To solve the overfitting problem, the dropout method is utilized to randomly drop neurons in the network, improving the generalization ability and training speed of model.\[45\] With the dropout technique, the neurons from in the neural network are randomly dropped, addressing the overfitting problem more efficiently during training. Neurons with all incoming and outgoing connections are temporarily removed from the network. The neuron is temporarily removed from the network along with all its incoming and outgoing connections. Each neuron is retained according to a specific fixed probability \( p \). The dropout layer is placed between the two fully connected layers. Consequently, a “thinned” network from origin neural network is obtained with dropout technique applied. The new network consists of all the neurons that survive in dropout process, being less sensitive to the specific weights of neurons.

**RMSprop algorithm**

The RMSprop technique is utilized to train the deep-learning neural network. Compared to other optimization methods, the RMSprop technique further improves convergence speed and convergence character. The update of the network parameters weight \( W \) and bias \( b \) can be described as:

\[
J(W, b) = \frac{1}{m} \sum_{j=1}^{m} \left( \hat{y}_j - y_j \right)^2 \quad \text{(Equation 18)}
\]

\[
S_1^t = \beta_1 S_1^{t-1} + (1 - \beta_1) \frac{\partial^2 J^{t-1}}{\partial W^2} \quad \text{(Equation 19)}
\]

\[
S_2^t = \beta_2 S_2^{t-1} + (1 - \beta_2) \frac{\partial^2 J^{t-1}}{\partial b^2} \quad \text{(Equation 20)}
\]

\[
W^t = W^{t-1} - \alpha \frac{\partial J^{t-1}}{\partial W} \sqrt{S_1^t} \quad \text{(Equation 21)}
\]

\[
b^t = b^{t-1} - \alpha \frac{\partial J^{t-1}}{\partial b} \sqrt{S_2^t} \quad \text{(Equation 22)}
\]

where \( J(\cdot) \) represents the cost function of the LSTM NN, \( t \) the number of training iteration, \( y \) the real value, \( \hat{y} \) the predicted value respectively, \( \beta \) the update coefficient of \( S \), and \( \alpha \) the learning rate.