Second-life Lithium-ion batteries: A chemistry-agnostic and scalable health estimation algorithm

Aki Takahashi, Anirudh Allam, and Simona Onori

Abstract

Battery state of health is an essential metric for diagnosing battery degradation during testing and operation. While many unique measurements are possible in the design phase, for practical applications often only temperature, voltage and current sensing are accessible. This paper presents a novel combination of machine learning techniques to produce accurate predictions significantly faster than standard Gaussian processes. The data-driven approach uses feature generation with simple mathematics, feature filtering, and bagging, which is validated with publicly available aging datasets of more than 200 cells with slow and fast charging, across different cathode chemistries, and for various operating conditions. Based on multiple training-test partitions, average and median state of health prediction root mean square error (RMSE) is found to be less than 1.48% and 1.27%, respectively, with a limited amount of input data, showing the capability of the approach even when input data and time are limiting factors. The process developed in this paper has direct applicability to today’s incumbent open challenge of assessing retired batteries on the basis of their residual health, and therefore nominal remaining useful life, to allow fast classification for second-life reutilization.

Introduction

Lithium-ion batteries are a powerful energy storage technology used in a wide variety of industries due to superior energy and power density among available electrochemical devices, as well as their cycling capability. However, they are prone to losses in performance over time due to multiple causes of deterioration, including loss of active material (LAM), corrosion, passivation, lithium plating, and solid electrolyte interphase layer growth [1].

It is therefore critical for users and battery manufacturers to be able to predict this deterioration accurately so that the battery can be safely utilized in later applications. One such critical application is the repurposing of retired batteries for grid-scale storage operation, such as peak-shaving or frequency regulation [33]. According to a McKinsey report, the supply of second-life EV batteries could surpass 100 gigawatt-hours per year by 2030, with the potential to meet half of the forecast global demand for utility-scale energy storage in that year [2]. One implication of such prediction is that a significant number of second-life EV batteries will need to be screened, in the near future, for accurate repurposing. This calls for a scalable and fast estimation algorithm of battery deterioration.

There are multiple approaches for estimating the battery health indicators (HIs), such as impedance, capacity, state of power, which express a battery’s deterioration metric based on a relative measure. Such approaches are based on models that fall in one of the following categories: equivalent circuit model (ECM) [35], physics-based [34], and data-driven models [3]. For example, thevenin equivalent circuits are often employed to model the fast dynamics of battery cycling [4] and equipped with empirical aging models [36]. Physics-based models use underlying electrochemical principles to model the battery, hence they have higher accuracy than ECs. At the same time, due to the complex nature of the coupled physical principles, the tradeoff between accuracy and computational burden becomes significant [5].

Data-driven approaches have shown promise in recent works. Once data is generated, a machine learning or statistical approach is employed to make predictions on data unseen by the model [6, 7, 8]. Examples include neural networks, where models independently find relationships between degradation indicators and battery health [9, 10]. Support vector regression and random forests are also used, where differences between features are defined in order to learn the relationship between features and the response [11, 12]. Gaussian process regression (GPR), a nonparametric approach, is an increasingly popular method because it is interpretable and addresses prediction uncertainty through confidence bounds [6, 13, 14]. There are different approaches to GPR, namely, where training is done on some early cycles and predictions are made there after [15, 16], or where models are trained on separate cells and then predictions are made on test cells [14, 17]. The former approach assumes that the battery undergoes similar processes in both early and later cycles, making it an inappropriate approach for repurposing applications. On the other hand, with the latter
approach the prediction algorithm can be extrapolated to cells with similar operating conditions. However, currently there is lack of a universal approach to battery health assessment and life prediction due to the nonlinear nature of battery degradation dynamics, which leads to the need for predicting cell life probabilistically and efficiently, regardless of cell chemistry [7, 18, 19].

In this paper, a data-driven approach which combines ensemble methods with GPR is proposed to quickly predict overall battery capacity. The model uses novel features from voltage and current information in a limited window of the charge profile. Time is not used explicitly, which, while valuable [15] may not be practical when the battery undergoes incomplete charging and discharging processes. Bagging is a facet of the ensemble learning, which has been used for battery state of health (SOH) prediction, but the models are mainly trained on early aging data for individual cells [8]. This single cell approach allows for models that are well calibrated to one particular cell, but adaptability is sacrificed, which is important in applications where many cells are required to be estimated, such as second life applications.

Significant contributions of this work are as follows: 1) an ensemble-based GPR model is proposed to quickly and accurately predict overall battery SOH, 2) novel features from voltage and current measurement that can be collected in as few as 2 minutes are proposed, 3) the model is tested on multiple datasets of lithium ion batteries, each with different experimentation and aging processes, thereby showing the wide applicability of the model. In addition, the model’s modular nature allows for easy customization of data generation, selection, and prediction.

### Results and Discussion

#### Battery HIs

Health indicators during the battery’s entire lifetime include, SOHₐ and SOHₑ which, in this work, is defined as a ratio of the battery’s current maximum capacity or energy relative to the maximum capacity or energy, respectively, measured at the beginning of life. They are defined as:

| Dataset Source | Cathode | Form factor | Nominal capacity | Temperature | Aging Protocol | Charging Current | Voltage range | Number of cells |
|----------------|---------|-------------|------------------|-------------|----------------|------------------|---------------|-----------------|
| [18] NMC       | Pouch   | 740mAh      | 40 °C            | ARTEMIS     | 1 C            | 2.7 ~ 4.2        | 8             |
| [19] LCO       | 18650   | 2.1 Ah      | Room temperature | Random      | 1 C            | 3.2 ~ 4.2        | 20            |
| [7] LFP        | 18650   | 1.1 Ah      | 30 °C            | 2 step CC-CV discharge | 1 ~ 6 C | 2 ~ 3.6        | 124           |
| [22] LFP       | 18650   | 1.1 Ah      | 30 °C            | 4 step CC-CV discharge | 4 ~ 8 C | 2 ~ 3.6        | 45            |

Table 1: Summary of different datasets and their experimental conditions.
SOH$_C(t) = \frac{Q(t)}{Q_{nom}} \cdot 100\% \quad (3)
\[
SOH_E(t) = \frac{E(t)}{E_{nom}} \cdot 100\% \quad (4)
\]

Where $Q(t)$ and $E(t)$ are the capacity and energy at time $t$. While a deterioration of 20% in SOH$_C$ is the industry standard for battery end of life, SOH$_E$ is supplemented with changes in the battery voltage.

As the battery is used, internal resistance increases due to the aging mechanisms, resulting in a shift of the capacity-voltage curve over time [20]. Thus, SOH$_E$ deterioration can also be used to identify the increase in resistance of the cell.

**Data Processing**

Datasets from online repositories were cleaned and used in this work (see Experimental Procedures), which is summarized in Table 1. A comparison between the battery health indicators, SOH$_C$ and SOH$_E$, is shown in the Fig. S1 found in the Appendix. The trend for all three datasets is quite different, likely due to differing conditions as outlined in Table 1. However, there is a strong linear relationship between the two HIs, so subsequent analysis will be conducted using the capacity-based SOH, SOH$_C$, simply denoted SOH.

Feature generation is done on the charge cycles since constant current – constant voltage (CC-CV) data is consistently available, compared to the discharge cycle which is heavily dependent on the intended application. The CC and CV regimes are significant regions where deterioration is highly observed. For the CC process, observations are limited to the upper half of the operating voltage. This is because degradation is most visible in said regions, as shown by the changing signals as cycles increase in Fig. 1. At the same time, we use a segment of the protocol for the feature generation, which is significant for reducing measurement time, and addressing issues when the battery is charged from a nonzero SOC. Once data is collected, it is processed by removing erroneous measurements and then interpolated (see Experimental Procedures) to get the necessary values at constant frequency for feature generation.

Fig. 1 illustrates the voltage vs. time and current vs. time curves over many cycles of a representative cell in the LFP dataset [7]. The datasets for other chemistries show similar trends over different time ranges due to the differing $C$ rate. Current measurements are also taken during CV at the known upper voltage limit. For the NMC dataset, the cells did not undergo CV, hence CV is not considered for analysis. The cutoffs are summarized in Supplementary Table 1.
The sampling rate for the features is an important design parameter since excessive data collection is computationally intensive. Battery voltage and current dynamics will also depend on C-rate, and some applications require frequent sampling [23]. For example, it is not necessary to sample at 1Hz for a charging process at 1C, since the change in the voltage and current curves occur slowly over an hour. Hence, the sampling rate varies depending on the time range that the datasets are charged for. The NMC and LCO datasets have >1 hour charge durations due to their low current, hence we take either voltage or current measurements every 10 seconds to get sufficient data until end of CC or CV. As for the LFP dataset, we take measurements every two seconds in a 30 second window for CC and 60 seconds for CV. The C-rate is higher, so measurements are only needed for a small amount of time to observe deterioration. This also allows us to demonstrate the capability of this approach in both slow and fast charge regimes, as well as a limited window of measurements.

For each of CC and CV, 7 simple statistical measures are used to characterize the data, summarized in Supplementary Table 2. The statistics are applied to voltage during CC and current during CV. The main benefit of using said statistics is their simplicity in calculation, and lack of reliance on specific measurements of time. In addition, they do not rely on derivatives, as seen in incremental capacity and differential voltage analysis, which can introduce noise without using a slow characterization procedure [24]. Lastly, the features are normalized by subtracting the same metric calculated at the start of life. This is useful for keeping consistent magnitudes for the cell features.

**Feature Selection**

Once the features are generated, a feature selection technique is used to eliminate redundant and noisy features. This reduces the dimensionality of the model, and allows for efficient and accurate predictions of SOH. There are a variety of selection techniques, such as filtering, wrapping, and fusion, with each varying in purpose and complexity [25].

Here, we use the fastest filter method via some correlation coefficient, such as Pearson or Spearman, to quickly determine which features correlate best with the predicted variable. The Pearson correlation coefficient assumes a Gaussian distribution for the vectors and a linear relationship between two variables. This assumption is sometimes not true due to the nonlinear nature of capacity degradation. Hence, the Spearman coefficient is found to be more suitable and therefore used. The equation is as follows:

\[
\rho = \frac{\sum_{i=1}^{n}(x_i - \bar{x})(y_i - \bar{y})}{\sqrt{\sum_{i=1}^{n}(x_i - \bar{x})^2 \sum_{i=1}^{n}(y_i - \bar{y})^2}}
\]  

(1)
where $i$ is the $i$-th value in the corresponding feature vector, $x$ is the feature, $\bar{x}$ is the average of the feature, $y$ is the response, $\bar{y}$ is the corresponding average response. This expression looks for a monotonic relationship between features and responses, meaning the magnitude of $\rho$ increases as the response either increases or decreases as the feature increases. Once the magnitude of $\rho$ was found for all features, we select 10 features with highest value, while also looking for dissimilar features ($|\rho| < 0.8$ between features), as done in [17]. An illustration is shown in Fig 2. An example of the relationship between features and SOH for representative cells is shown in the Appendix Fig. S3, S4, and S5.

**Scalable Gaussian Process Regression**

GPR is a nonparametric probabilistic algorithm used to make predictions based on Gaussian distributions of features and response [26]. Specifically, the Gaussian process regression (GPR) is defined as:

$$GPR (M(x), k(x))$$ (2)

where $M(x)$ is the mean function and $k(x)$ is the covariance function of the feature and response. If each set of the inputs has a joint Gaussian distribution, then the whole set of inputs forms a joint Gaussian distribution. This means that the predicted response has a distribution, with $M$ and $k$ defining the response and its uncertainty. Further details are provided in Experimental Procedures.

While GPR models are effective, they suffer from scalability, since for model training there are $O(n^3)$ and $O(n^2)$ computation and storage requirements in big-O notation, while the cost of a single prediction is $O(n)$, where $n$ is the number of datapoints being trained on. This is a particular problem for second-life applications where large number of batteries need to be processed. To mitigate this, one approach is to use bagging (bootstrap aggregating), an ensemble learning technique used mainly for random forest algorithms. However, this approach is capable of being applied to many different machine learning algorithms, including GPR [27]. From a large dataset, $m$ different bags of size $n$ are created by randomly sampling from the dataset with replacement, meaning some examples can be selected more than once. Once the $m$ sampled datasets are created, a GPR model is trained on each dataset to create $m$ models. For subsequent predictions then, these models are combined using some aggregating technique, such as a simple average or weighted average of the predicted output. It should be noted that this is different from Bayesian Committee Machines, which is another method used for scalable GPs, where all of the training data is split into subsets instead of random sampling with replacement [28].

Bagging is attractive for its stability and reduction of variance, since it does not rely on a single model, whose error can increase quickly with degradation if training data does not relate to the testing data. It also allows for parallel computing since each of the bagged models undergo training and prediction separately. In the case of battery degradation, it also means we can create “pseudo-cells” that are not aware of single cell-specific aging mechanisms and are able to use a section of the data for prediction. Each of the GPR models can make their own predictions, and when combined, their net performance improves noticeably [27]. As for the prediction, we use a weighted average to account for the predictive capability of each of the bags (see Experimental Procedures).

| Training on large amount of data (N = 10000) | Training on bagged data (m = 50, n = 200) |
|-------------------------------------------|-------------------------------------------|
| 51 seconds                                | 11 seconds                                |

Table 2: Comparison in seconds of training time for the LFP set when the same amount of data is used. Training time is reduced by a factor of roughly 5 due to the smaller bag size.

Computational complexity needed to train the bagged model can be reduced significantly by using bags smaller than the whole dataset, as well as using parallel computing features. A demonstration is shown in Table 2, where the training time is compared on partitions of the LFP dataset using a i7-9750H CPU @ 2.60GHz and the parallel computing feature in MATLAB with 6 workers. In general, the bagged approach reduces the amount of training time.
since there is no need to optimize the hyperparameters of the GPR on a large feature set. With bagging, computational complexity becomes $O(mn^3)$, meaning that with smaller selection of $m$ and $n$ GPRs can be simplified exponentially with strong accuracy [29]. Specifically, the data used by the models can be compared using the factor reduction of data (FRD) metric as follows:

$$FRD = \frac{N}{mn}$$  \hspace{1cm} (3)

For constant $N$, the original number of datapoints, the FRD can be increased by changing $m$ and $n$. A larger FRD leads to a significant increase in model speed for training and prediction when combined with parallel computing.

**Model Performance**

To measure model performance, an approximate 70-30 training-test split is randomly created based on individual cells. While out of bag error is commonly associated with bagging techniques, testing on the entire battery lifetime for individual cells should be preferred over unrelated datapoints in the context of battery aging and longitudinal data in general [30].

The proposed approach allows to analyze when models succeed and fail at predicting battery SOH, since predictions are made for specific cells. The actual split is slightly different from 70-30 based on the divisibility of the number of cells for each dataset. Feature selection is conducted on the training set and features with low correlation are eliminated. Then, the bagged models are created and trained. Finally, the models perform predictions on all available test data with confidence intervals, and the results are aggregated using an averaging rule. An example of the prediction timeline is shown in Fig. 3. This process is repeated several times for smaller datasets to be able to compare a similar number of tested cells. To evaluate model performance, we use root mean square percent error (RMSPE) and mean percent error (MPE) (see Experimental Procedures). This combination allows to report model performance based on the presence of outliers and overall model performance. Furthermore, we use a calibration score (CS) to measure model uncertainty [13]. Formal definitions are described in the Experimental Procedures.

**SOH Estimation Results**

For all datasets $m$ was initially selected to be half of the number of training cells, with sample size $n$ being close to the average number of characterization cycles for each cell, then modifications were made based on performance. The factor reduction in training data is a rough estimate derived from a typical training data split.

Specific hyperparameters are explained in each section of the following sections and summarized in Table 3. In addition, for all datasets we repeat the process shown in Fig. 3 multiple times to get 300 test cells for all datasets to
compare amongst them using the same amount of test data. All results are shown in Fig. 4,5 and Table 4. Fig. 4 describes the distribution of the error of individual SOH predictions, while Fig. 5 compiles the metrics of individual cells into boxplots. Boxplots in Fig. 5 are used instead of conventional histograms to demonstrate performance on individual cells in addition to compare the different chemistries. Error mean and median are reported to describe the distribution, as well as an average CS score. Two sample cells, one with low error and another with high error are reported for a fair demonstration of model performance. This provides a baseline for a comprehensive analysis of the model.

**Table 3:** $m$ and $n$ used in model implementation for each cathode chemistry. Factor reduction in training data is found by dividing the typical amount of training data by the number of examples used in the bagged approach. This leads to an exponential decrease in training time.

|       | $m$ (number of models) | $n$ (sample size) | Factor reduction in training data |
|-------|-----------------------|-------------------|----------------------------------|
| NMC   | 3                     | 20                | 8                                |
| LCO   | 7                     | 30                | 2                                |
| LFP   | 20                    | 200               | 23                               |

**Table 4:** Overall % error for every cell in the testing cells based on the bagged approach. All datasets are tested to have a total of 300 test cells based on randomized bags.

|       | NMC | LCO | LFP |
|-------|-----|-----|-----|
| RMSPEmedian (%) | 0.3384 | 1.099 | 1.266 |
| RMSPEmean (%)    | 0.2464 | 1.277 | 1.475 |
| MPEmedian (%)    | 0.286  | 0.839 | 0.907 |
| MPEmean (%)      | 0.28486 | 0.925 | 1.286 |
| CSmean           | 66    | 91   | 72   |

Fig. 4: Actual vs. predicted SOH for all individual training and test datapoints for (a) NMC, (b) LCO, (c) LFP. 20 bins are used to show the overall distribution of each dataset.
NMC Dataset

This dataset has the least cells; however, the 70-30 split is still valid with 6 cells used for training and 2 cells for testing in each iteration. 3 bags were created, each with 20 sample points, to reduce the training data to approximately 1/8 of its original size for each iteration and hence exponentially faster. Since there are not many cells the process is repeated 150 times to get a total of 300 estimates. The results in Fig. 4 and 5 show that the bagged GPR prediction performs well, with the lowest median MPE and RMSPE out of the three datasets. The boxplots show a low RMSPE and MPE for all 300 cells overall with its rightward skew, which suggests that the GPR bagged model was able to successfully predict the degradation for the NMC cells. Some of the higher RMSPE errors are likely due to the sudden drop in SOH past 50% in one of the cells; MPE is significantly lower since the effect of this single anomaly is small. The prediction interval is narrow for the representative cell even though the maxima are used for the bounds, which is likely due to the very high correlation coefficient (> .95) between the CC features and SOH for individual cells. This is shown by the overconfident CS\textsubscript{mean} which was less than .95. It should be reiterated that these cells do not undergo a CV regime during the charging process, meaning that while we look for the 10 best features there are only 7 features used. Regardless of the reduction of features, the bagged model was able to make accurate predictions, starting from a voltage of 3.45 V.

LCO Dataset

14 cells were selected for training and 6 were used for testing in each iteration of prediction. Predictions are made to the end of available data. Based on the number of cells and number of reference charges conducted, 7 bags with sample size 30 were selected to reduce the training data. 300 predictions are made based on 50 iterations. Like the NMC set, the median RMSPE and MPE are low as shown in Fig. 5a and 5b, with slightly higher mean values as expected due to the skew towards higher error shown in the boxplot. In this dataset the sum of the voltage values was an important feature within the observed CC range of 3.6 to 4.2V. This may be due to the long charge characterization, in that the CC regime was significantly longer and hence the feature generation captured more voltage points to add up in early cycles of the battery. The LCO dataset also provided the best CS, which was only slightly overconfident. In addition, this dataset shows the capability of SOH prediction far past the standard end of life of 80%, as data is available for up to 50% SOH.

LFP Dataset

Since this dataset has the most data the amount is reduced accordingly through bagging without sacrificing significant accuracy. Four cells were anomalous in measurements, so they were excluded from analysis, leaving 165 cells. A training test split uses 115 cells for training and 50 cells for testing. Training data was reduced to less than 1/20 of a typical training set size. For each test set there are 50 cells, so with 6 iterations there are 300 predictions made, whose error is shown in Fig. 5. Since the boxplots are positively skewed, the median and mean are reported, with the median RMSPE being slightly lower than the average. The median MPE was under 1%, meaning that all three datasets were able to achieve under 1% error for the median MPE. While this error is relatively high compared to the other two datasets, the representative cell shows that the actual SOH is within the confidence interval throughout the entire aging process. In addition, there are no significant outliers in the error, suggesting the stability of the bagging approach. The
higher error compared to the other two cell chemistries may be due to the wide range of days that the cells lasted for as well as the limited window of time that measurements were made on. However, with fast charging becoming more important than ever, it is important to be able to extract important features as shown with this dataset within a small amount of time.

**Effect of \( m \) and \( n \)**

In bagging techniques, there is a convention to choose \( n \) to be the same size as the training data whereas \( m \) is arbitrarily selected [30]. However, for GPs it is impractical to use large datasets due to the increasing training time and storage space, as well as the diminishing improvements in accuracy [29]. Hence, we analyze the effect of changing \( m \) and \( n \) in a set domain to see how much data can be eliminated. First, the sample size \( n \) is selected, then create \( M \) number of bags, where \( M \) is the greatest number of bags used. Then, we train all \( M \) models, and use small batches of size \( m \) out of the \( M \) models for prediction. This methodology allows for comparisons between performance of bagged models with the same \( n \) but different \( m \), as the same subsamples are used and performance is measured based on \( m \), not random sampling. The process is repeated 10 times for each dataset.
Since the datasets are of different sizes, the number of bags and sample size are different, which is outlined in Supplementary Table 3. The values are based on the diminishing changes in performance. The results are observed in Fig. 6. Note that the mean MPE and RMSPE are presented, since while the median may be appropriate based on the skewed boxplot distributions, the mean is affected by outliers and hence it is possible to observe the stability better.

In general, it can be observed in Fig. 6 that increasing \( m \) and \( n \) decreases the average RMSPE and MPE. Interestingly, the NMC dataset had worse predictions when the bags are larger, albeit the increase in error is not significant. This may be due to the high predictive capability of the CC features. Including more data reaffirms the linear correlation between the features and SOH, and hence the predictions are made overconfidently. Increasing the number of bags past 10 did not have significant improvements, meaning that it is possible to make good predictions with as little as 10 sample points each in 10 bags, which reduces the amount of training data by a factor of 4 and exponentially decreases computational complexity.

With the LCO dataset, the average RMSPE and MPE error quickly decrease as the number of bags and sample size increase. The data needed to make better predictions was relatively more than the NMC dataset, which is to be expected due to the variable aging procedures. The average MPE was below 0.8% and RMSPE was close to 1% even with a relatively small \( m \) and \( n \) of 10 and 80, for example, with less memory usage compared to using all data (0.802 megabytes vs. 1.13 megabytes based on data from 14 cells).

Fig. 6c highlights the capability to reduce computational complexity where there is lots of aging data. It is impractical and unnecessary to use all training data, which was typically \( n = 90000 \), to train a single GPR model. Using many bags and a small sample size, the bagged approach can make good predictions with error less than 1.5% in average RMSPE and 1.18% in average MPE past \( n = 850 \) and \( m = 20 \), which is much more practical to use.

**Discussion**

The proposed ML algorithm showed satisfactory results to predictions based on pure current and voltage charging data from datasets of different chemistries and aging mechanisms. With scalable SOH estimation becoming more relevant, the used features demonstrate the capability of making predictions within a 30 second span on fast-charge voltage data and 1 minute of current data, as well as traditional long characterization cycles. It should be noted that while we start measurements from the upper half of the operating voltage, this voltage range can be adjusted based on the desired operating conditions. The feature generation process should work as intended regardless, provided that within the selected region changes in voltage and current are visible over time. This is because the features characterize the voltage and current curve in different ways, such as CC or CV occurring over a longer period as the cell ages (sum) or reaching a higher voltage value faster (median). This also means that if the batteries undergo fast charge, the window of measurements can be made much smaller since the changes are more apparent in a shorter amount of time between individual charging procedures.

The LCO and NMC dataset had higher degrees of success, likely due to the relatively similar degradation over time between cells, but nevertheless highlights the ability for quick, safe, and accurate predictions of SOH for a multitude of cells with a single model, which potentially expands to pack level SOH estimation. Since data-driven approaches depend on the quality of the training data, one potential solution for accurate estimation is to cycle batteries under different operating conditions, “package” them into small GPR models, then switch out and combine models via bagging depending on their predictive capability [32]. This will be relevant for practical applications, where operating conditions of the batteries could vary significantly depending on driving patterns and local environment. A consideration to be made also is the tradeoff with uncertainty. Using similar data can cause overconfidence in models, while on the other hand using not enough data will lead to loss in accuracies. The search space for \( m \) and \( n \) provides guidance on this tradeoff, since it is possible to easily observe the change in performance metrics.
Ultimately, the bagged approach can save on memory and processing power even with a rich amount of data. This can be expanded to practical applications due to the way the data is collected on the charging curve, and serves as an effective diagnostic technique for second-life applications. We acknowledge that there are other ways to scale Gaussian Processes in the literature, such as the Bayesian Committee Machine or approximating the kernel function, and these introduce interesting areas that can be explored in the context of battery degradation [28,29].

Conclusion

Battery data is becoming more important and abundant than ever, therefore there is a need for a fast and accurate way to diagnose the SOH of many batteries, especially for repurposing purposes. The proposed novel method consists of: using simple statistics to describe a small window of voltage and current data, fast feature filtering, and bagging multiple GPR models. The approach was found to be an efficient and accurate method of predicting battery SOH due to each modularized step. Specifically, using as little as 1 minute of current data and 30 seconds of voltage data and less than 1/20 of the training data, the proposed approach was able to predict battery SOH for 300 test cells with under 1.5% average RMSPE. The approach worked well for data with low C-rate characterization as well, achieving about 0.5% and 1.27% average RMSPE for NMC and LCO batteries respectively. This suggests the applicability in both fast and slow charging application, as well as for different form factors, temperature, cathode chemistry, and other operating conditions. Especially with fast charge, the diagnosis time can be significantly reduced through this approach due to the increased change in voltage over time.

Finally, this work has highlighted the opportunity and considerations required for a ML based algorithm to estimate battery health for repurposing applications, solely based on a limited window of voltage and current measurements. For future works, the scalability of data-driven models, in addition to accurate and safe predictions is a key design parameter for transitioning models to industrial applications and achieving sustainability goals.

The approach presented in this paper shows the feasibility to use data-driven models over portion of CC-CV data to make quick and accurate assessments of battery health. This can ultimately be directly applied to the problem of retired batteries classification for repurposing and reutilization aims.

Experimental Procedures

Datasets

Datasets from online repositories were used in this work. Each of the datasets used unique aging experiments and different battery chemistries – namely, LFP, NMC, and LCO. Aging data used in this paper for all cells is shown in the Appendix and summarized in Table 1.

1. Nickel Manganese Cobalt Oxide (NMC)

This dataset used 8 Kokam SLPB533459H4 NMC cells, which aged via the ARTEMIS driving schedule [21]. Characteristic data is taken for every 100 cycles with a 1C full charge-discharge cycle and a constant-current OCV test. The discharge capacity data is available hence it is used for SOH calculations.

2. Lithium Cobalt Oxide (LCO)

The LCO dataset featured in [19] uses statistically random discharge for battery deterioration. The cells used were LG Chem. 18650 LCO cells. The cells were divided into multiple groups of 4, each undergoing a unique, randomized charging and discharging procedure at room temperature. A characteristic charge-discharge cycle at 2A took place periodically, allowing for a comparison between aging procedures. Capacity data is unavailable so the discharge curve is integrated to obtain SOH measures.

3. Lithium Iron Phosphate (LFP)

In this group of cells, 124 commercial high-power LFP A123 APR18650M1A cells were aged via a full two-step fast charging cycle and 4C discharge [7]. Another batch of 45 cells from the same manufacturer underwent a 10 minute 4-step fast charge cycle and 4C discharge [22]. Once cells reached 80% nominal capacity a 1C charging regime followed
by constant voltage (CV) charge was used to fully charge the cells in both datasets. There are three different batches of cells in [7], each differing by the amount of rest taken between charging and discharging phases. Discharge capacity data is available, so we use these measurements as the expected response.

**Data Preprocessing**

For data processing on MATLAB, erroneous measurements, such as battery voltage exceeding cutoff values, were first removed. An interpolation scheme is then used on the voltage and current charging curves to obtain regular measurements. While battery data is dynamic, because of the frequency ranges used (2 – 10 seconds) for the ML model the data required is less than the lab data frequencies, which is often measured less than every second. Thus, a simple linear interpolation was enough for collecting accurate measurements of battery voltage and current. Measurements are then collected at the desired frequency based on the starting voltage or current value.

**Gaussian Process Regression Details**

The Matern 5/2 covariance function is used for all three datasets since it is able to adapt to different smoothness throughout the regression problem [14, 26]. The function is as follows:

\[
    k(x, x') = \sigma_f^2 \frac{2^{1-v}}{\Gamma(v)} \left( \frac{\sqrt{2v} \frac{x - x'}{\xi}}{v} \right)^v K_v\left(\frac{\sqrt{2v} \frac{x - x'}{\xi}}{v}\right)
\]

where \(\sigma_f^2\) is the noise variance, \(\Gamma\) is the gamma function, \(K_v\) is the modified Bessel function, \(\xi\) is the length scale, and \(v = 5/2\) which simplifies the expression considerably. The hyperparameters that need to be optimized are the length scale, a measure that outlines how far extrapolations can be made, and the noise variance, which determines the spread of the joint distribution. Essentially, optimizing the set of hyperparameters allows the GPR model to fit a set of input variables \(X\) to model a Gaussian with respect to the response \(y\). The hyperparameters are optimized by minimizing a negative log marginal likelihood function, which allows for automatic tradeoff between bias and variance. The equation is as follows:

\[
    L = -\frac{1}{2} \log(\det(k_f + \sigma_n^2 I)) - \frac{1}{2} y^T [K_f + \sigma_n^2 I]^{-1} y - \frac{N}{2} \log(2\pi)
\]

where \(k_f\) is the selected covariance function, \(\sigma_n^2\) is the noise variance, and \(I\) is the identity matrix.

**Predictions with Bootstrap Aggregating**

For bagging, the predictions of each model must be combined to generate a prediction. While weightless aggregation is possible, it has been shown that a weighted prediction is often better [27]. For this work, predicted responses are weighted based on the standard deviation. The weight function is as follows:

\[
    w_a = \frac{1}{\sigma_a}
\]

where \(a\) is the \(a\)-th GPR model in the \(m\) model set for a particular prediction, \(\sigma\) is the associated error standard deviation. In general, this means that a more unconfident prediction is punished more, and it has been shown that a weighted average performs better [27]. The standard deviation is used instead of the variance since the variance more heavily favors or punishes individual models, which leads to significantly overconfident predictions with large \(m\) and \(n\) [29]. This expression is multiplied with the corresponding prediction to establish a weighted average, which also has a weighted standard deviation as follows:

\[
    y_{pred} = \frac{\sum_{a=1}^{m} w_a y_a}{\sum_{a=1}^{m} w_a}
\]
\[ \sigma_{\text{pred}} = \sqrt{\frac{Z \sum_{a=1}^{m} w_a (y_a - y_{\text{pred}})^2}{(Z - 1) \sum_{a=1}^{m} w_a}} \]  

where \( y_{\text{pred}} \) is the aggregated SOH prediction, \( y_a \) is the prediction made by the a-th model out of the m-model set, \( \sigma_{\text{pred}} \) is the predicted standard deviation and \( Z \) is the number of nonzero weights (which is almost always the same as \( m \) in our implementation, but is included for greater generalizability). The confidence interval will be 95%, or 1.96 * \( \sigma_{\text{pred}} \) from the predicted SOH. This allows this bagged approach to quickly make predictions for many cells and with an expression of uncertainty for individual cells.

**Model Performance Metrics**

Formally, RMSPE and MPE are defined as follows:

\[
\text{RMSPE} \left( \% \right) = \frac{1}{c} \sum_{j=1}^{c} \left( \frac{y_{\text{exp},j} - y_{\text{pred},j}}{y_{\text{pred},j}} \right)^2 \times 100\%
\]

\[
\text{MPE} \left( \% \right) = \frac{1}{c} \sum_{j=1}^{c} \left| \frac{y_{\text{exp},j} - y_{\text{pred},j}}{y_{\text{pred},j}} - 1 \right| \times 100\% / x
\]

where \( j \) is the j-th datapoint in the \( c \) characteristic cycles of a single cell, \( y_{\text{exp}} \) is the expected SOH from the discharge data and \( y_{\text{pred}} \) is the weighted average of the predicted SOH. The calibration score measures the reliability of the standard deviation, and the definition is as follows:

\[
CS = \left( \frac{1}{x} \sum_{i=1}^{x} |y_{\text{exp}} - y_{\text{pred}}| < 1.96 \sigma_{\text{predicted}} \right)
\]

It can be said that we are 95% confident that \( y_{\text{exp}} \) lies inside the interval predicted by the GPR, with a lower percentage suggesting overconfidence and higher percentage suggesting underconfidence. The CS measure should therefore be as close as possible to 0.95 to demonstrate reliability.
References

[1] Vetter, J., Novák, P., Wagner, M.R., Veit, C., Möller, K., Besenhard, J.O., Winter, M., Wohlfahrt-Mehrens, M., Vogler, C., & Hammouche, A. (2005). Ageing mechanisms in lithium-ion batteries. *Journal of Power Sources, 147*, 269-281.

[2] H., Engel. *McKinsey Center for Future Mobility, Global Editorial Services*, 2019

[3] Ng, M., Zhao, J., Yan, Q., Conduit, G.J., & Seh, Z.W. (2020). Predicting the state of charge and health of batteries using data-driven machine learning. *Nature Machine Intelligence, 2*, 161-170.

[4] Li, K., Wei, F., Tseng, K.J., & Soong, B.H. (2018). A Practical Lithium-Ion Battery Model for State of Energy and Voltage Responses Prediction Incorporating Temperature and Ageing Effects. *IEEE Transactions on Industrial Electronics, 65*, 6696-6708.

[5] Bizeray, A.M., Kim, J., Duncan, S.R., & Howey, D.A. (2019). Identifiability and Parameter Estimation of the Single Particle Lithium-Ion Battery Model. *IEEE Transactions on Control Systems Technology, 27*, 1862-1877.

[6] Hu, C., Jain, G., Tamirisa, P.A., & Gorka, T. (2014). Method for estimating capacity and predicting remaining useful life of lithium-ion battery. *2014 International Conference on Prognostics and Health Management, 1*-8.

[7] Severson, K.A., Attia, P.M., Jin, N., Perkins, N., Jiang, B., Yang, Z., Chen, M.H., Aykol, M., Herring, P.K., Fragededakis, D., Bazant, M.Z., Harris, S.J., Chueh, W.C., & Braatz, R.D. (2019). Data-driven prediction of battery cycle life before capacity degradation. *Nature Energy, 4*, 383-391.

[8] Xing, Y., Ma, E.W., Tsui, K., & Pecht, M.G. (2013). An ensemble model for predicting the remaining useful performance of lithium-ion batteries. *Microelectron. Reliab.*, 53, 811-820.

[9] Yang, D., Wang, Y., Pan, R., Chen, R., & Chen, Z. (2017). A Neural Network Based State-of-Health Estimation of Lithium-ion Battery in Electric Vehicles ☆. *Energy Procedia, 105*, 2059-2064.

[10] Roman, D., Saxena, S., Robu, V., Pecht, M.G., & Flynn, D. (2021). Machine learning pipeline for battery state of health estimation. *Nat. Mach. Intell., 3*, 447-456.

[11] Mansouri, S.S., Karvelis, P.S., Georgoulas, G., & Nikolakopoulos, G. (2017). Remaining Useful Battery Life Prediction for UAVs based on Machine Learning. *IFAC-PapersOnLine, 50*, 4727-4732.

[12] Nuhic, A., Terzimehic, T., Soczka-Guth, T., Buchholz, M., & Dietmayer, K.C. (2013). Health diagnosis and remaining useful life prognostics of lithium-ion batteries using data-driven methods. *Journal of Power Sources, 239*, 680-688.

[13] Richardson, R.R., Birkl, C.R., Osborne, M.A., & Howey, D.A. (2019). Gaussian Process Regression for In Situ Capacity Estimation of Lithium-Ion Batteries. *IEEE Transactions on Industrial Informatics, 15*, 127-138.

[14] Richardson, R.R., Osborne, M.A., & Howey, D.A. (2017). Gaussian process regression for forecasting battery state of health. *Journal of Power Sources, 357*, 209-219.

[15] Yang, D., Zhang, X., Pan, R., Wang, Y., & Chen, Z. (2018). A novel Gaussian process regression model for state-of-health estimation of lithium-ion battery using charging curve. *Journal of Power Sources, 384*, 387-395.

[16] Yu, J. (2018). State of health prediction of lithium-ion batteries: Multiscale logic regression and Gaussian process regression ensemble. *Reliab. Eng. Syst. Saf.*, 174, 82-95.

[17] Greenbank, S., & Howey, D.A. (2021)."Automated feature extraction and selection for data-driven models of rapid battery capacity fade and end of life." *IEEE Transactions on Industrial Informatics*

[18] Birkl, C. (2017). Oxford Battery Degradation Dataset 1. University of Oxford.

[19] Bole, B., Kulkarni, C.S., & Daigle, M.J. (2014). Adaptation of an Electrochemistry-based Li-Ion Battery Model to Account for Deterioration Observed Under Randomized Use.
[20] Schweiger, H., Obeidi, O., Komesker, O., Raschke, A., Schiemann, M., Zehner, C., Gehnen, M., Keller, M., & Birke, P. (2010). Comparison of Several Methods for Determining the Internal Resistance of Lithium Ion Cells. *Sensors (Basel, Switzerland), 10*, 5604 - 5625.

[21] André, M. (2004). The ARTEMIS European driving cycles for measuring car pollutant emissions. *The Science of the total environment, 334-335*, 73-84.

[22] Attia, P.M., Grover, A., Jin, N., Severson, K.A., Markov, T., Liao, Y., Chen, M.H., Cheong, B., Perkins, N., Yang, Z., Herring, P.K., Aykol, M., Harris, S.J., Braatz, R.D., Ermon, S., & Chueh, W.C. (2020). Closed-loop optimization of fast-charging protocols for batteries with machine learning. *Nature, 578*, 397-402.

[23] Garche, J., & Jossen, A. (2000). Battery management systems (BMS) for increasing battery life time. *TELESCON 2000. Third International Telecommunications Energy Special Conference (IEEE Cat. No.00EX424)*, 81-88.

[24] Bloom, I.D., Jansen, A.N., Abraham, D.P., Knuth, J., Jones, S.A., Battaglia, V.S., & Henriksen, G.L. (2005). Differential voltage analyses of high-power, lithium-ion cells: 1. Technique and application. *Journal of Power Sources, 139*, 295-303.

[25] Hu, X., Che, Y., Lin, X., & Onori, S. (2021). Battery Health Prediction Using Fusion-Based Feature Selection and Machine Learning. *IEEE Transactions on Transportation Electrification, 7*, 382-398.

[26] Rasmussen, C.E., & Williams, C.K. (2009). Gaussian Processes for Machine Learning. *Adaptive computation and machine learning*.

[27] Chen, T., & Ren, J. (2009). Bagging for Gaussian process regression. *Neurocomputing, 72*, 1605-1610.

[28] Tresp, V. (2000). A Bayesian Committee Machine. *Neural Computation, 12*, 2719-2741.

[29] Liu, H., Ong, Y., Shen, X., & Cai, J. (2020). When Gaussian Process Meets Big Data: A Review of Scalable GPs. *IEEE Transactions on Neural Networks and Learning Systems, 31*, 4405-4423.

[30] Martínez-Muñoz, G., & Suárez, A. (2010). Out-of-bag estimation of the optimal sample size in bagging. *Pattern Recognit., 43*, 143-152.

[31] Weng, C., Cui, Y., Sun, J., & Peng, H. (2013). On-board state of health monitoring of lithium-ion batteries using incremental capacity analysis with support vector regression. *Journal of Power Sources, 235*, 36-44.

[32] Li, Y., Zou, C., Berecibar, M., Nanini-Maury, E., Chan, J.C., Van Den Bossche, P., Van Mierlo, J., & Omar, N. (2018). Random forest regression for online capacity estimation of lithium-ion batteries. *Applied Energy*.

[33] Moy, K., Lee, S., Harris, S., Onori, S. Design and validation of synthetic duty cycles for grid energy storage dispatch using lithium-ion batteries, *Advances in Applied Energy, Volume 4*, 2021

[34] Allam, A., Onori, S., "An Interconnected Observer for Concurrent Estimation of Bulk and Surface Concentration in Cathode and Anode of a Lithium-ion Battery". *IEEE Transactions on Industrial Electronics, pp. 7311 - 7321 Vol. 65, Issue: 9, 2018*

[35] Taborelli, C., Onori, S., Maes, S., Sveum, P., Al-Hallaj, S., Al-Khayat, N., "Advanced battery management system design for SOC/SOH estimation for e-bikes applications", *International Journal of Powertrains, Vol. 5, n. 4, 2016*

[36] Todeschini, F., Onori, S., Rizzoni, G., "An experimentally validated capacity degradation model for Li-ion batteries in PHEVs applications", *8th IFAC International Symposium on Fault Detection, Supervision and Safety of Technical Processes, Mexico, August 29-31, 2012*
Fig. S1: SOHc vs. time for a) NMC [18], b) LCO [19], c) LFP [7,22]. They are of different cell chemistries and operate on a different range of days as well as aging procedures.
Figure S2: Correlation and change in SOHc and SOHe for all cells for (a) NMC, b) LCO, c) LFP. A linear relationship can generally be observed for all three groups of cells, with a high correlation coefficient for all three datasets, irrespective of the testing operating conditions. In a), the linear fit is almost perfect, which suggests that SOHc and SOHe are interchangeable for this particular chemistry. In b), correlation is high, albeit lower than the other two chemistries. For c) it should be noted that in [7] the capacity-voltage feature used to predict capacity degradation was a measure of SOHe, which can be observed to decrease faster compared to SOHc. However, SOHe was not observed to deteriorate faster than SOHc in the other two datasets.
Supplementary Table 1: Values of upper, half, and lower voltage [V] and current [A] thresholds for each of the datasets used. The NMC cells do not undergo CV in the charge preceding the cycle, hence no current measurements are collected. The V window range starts from \( V_{\text{half}} \) to the specified condition, whereas the I window range starts from the start of CV to the specified condition.

| Statistics                  | Meaning                                      |
|-----------------------------|----------------------------------------------|
| Mean                        | Average over recorded values                 |
| Median                      | Central value of distribution                |
| Sum                         | Adding all recorded values                   |
| Standard Deviation          | Measure of spread with respect to mean       |
| Variance                    | Measure of spread                            |
| Kurtosis                    | Measure of deviation from normal distribution at extreme values |
| Interquartile range         | Measure of spread between 25% and 75% percentile |

Supplementary Table 2: The list of statistics applied on the voltage and current measurements. The selected statistics are chosen for their ease of calculation, and their definitions are briefly explained.
Figure S3: SOH vs. 7 features in Cell 4 of the NMC dataset [18]. Stronger relationships between the features and SOH were important for the model to learn the training data effectively and make strong predictions. There are no current statistics since the cells did not undergo a constant voltage portion.
Figure S4: SOH vs. 14 features in Cell 7 of the LCO dataset [19]. Stronger relationships between the features and SOH were important for the model to learn the training data effectively and make strong predictions.
Figure S5: SOH vs. 14 features in Cell 4 of the LFP dataset [7]. Stronger relationships between the features and SOH were important for the model to learn the training data effectively and make strong predictions.
| Dataset | $m$     | $n$     |
|---------|---------|---------|
| NMC     | 2 to 100| 10 to 150|
| LCO     | 2 to 50 | 10 to 150|
| LFP     | 2 to 80 | 50 to 1000|

Supplementary Table 3: The domain for $m$ and $n$ for each cell chemistry. The range is selected on the dataset size and diminishing improvements in the performance of the model.