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Useful Energy Prediction Model of a Lithium-ion Cell Operating on Various Duty Cycles

Damian Burzyński

Abstract — The paper deals with the subject of the prediction of useful energy during the cycling of a lithium-ion cell (LIC), using machine learning-based techniques. It was demonstrated that depending on the combination of cycling parameters, the useful energy (RUE_c) that can be transferred during a full cycle is variable, and also three different types of evolution of changes in RUE_c were identified. The paper presents a new non-parametric RUE_c prediction model based on Gaussian process regression. It was proven that the proposed methodology enables the RUE_c prediction for LICs discharged, above the depth of discharge, at a level of 70% with an acceptable error, which is confirmed for new load profiles. Furthermore, techniques associated with explainable artificial intelligence were applied, for the first time, to determine the significance of model input parameters – the variable importance method – and to determine the quantitative effect of individual model parameters (their reciprocal interaction) on RUE_c – the accumulated local effects model of the first and second order. Not only is the RUE_c prediction methodology presented in the paper characterised by high prediction accuracy when using small learning datasets, but it also shows high application potential in all kinds of battery management systems.

Index Terms— cycle life modelling, Gaussian mixture model, lithium-ion battery, machine learning, predictive models, useful energy prediction.

I. INTRODUCTION

IN recent years, it is possible to observe a rapid increase in the demand for equipment designed for electric energy storage. At present, it is estimated that the highest increase refers to the industry branch related to electromobility, where solutions based on lithium-ion batteries are currently dominant. It is predicted that by the end of 2030, the demand for batteries will increase ten times in this sector alone [1]-[2]. The prevailing share of lithium-ion batteries on the market follows primarily from their ability to transfer energy quickly and effectively, whereby this ability is successively increased owing to the dynamic development of electrochemical battery technology [3].

The key issue related to the operation of these storages is

their life. Degradation of lithium-ion cells (LICs) is a consequence of aging processes which take place during cycling, and also during the storage period [4]-[5]. Out of more than a dozen identified aging phenomena, lithium plating, formation, evolution and dissolution of the solid electrolyte interface (SEI), as well as electrolyte decomposition, particle gassing and corrosion of the current collectors are the pre-dominant contributors to the degradation process [6]. Without interference in the LIC structure, the effects of the aging processes may be determined through the loss in charge throughput and the increase in internal resistance (or impedance).

A. RELATED PAPERS

Based on the current state of knowledge, it can be concluded that in the case of LICs, usually separate degradation models are developed for cycling (cycle life) and storage period (calendar aging). Degradation models during cycling are multi-parameter models – their parameters include: values of discharge/charge current (I_d/I_{ch}), depth of discharge (DoD), cell temperature (T_c) or ambient temperature, as well as the charge or energy throughput (expressed most frequently as the number of full equivalent cycles – $FECs$). At this point, it must be emphasised that some of these parameters are correlated with each other and their effect on the amount of energy throughput of LICs is non-linear [7]. The above-mentioned facts make analysing and predicting the LICs' ability to transfer energy over the period of their lifetime highly complicated and require advanced methods. Regardless of the chemical composition of the cells, testing them is very time consuming. For this reason a more and more frequent practice is to conduct the testing under conditions which result in their accelerated degradation (e.g. under an increased load or at extreme temperatures), and then to predict their operational parameters under the assumed operating conditions [8]-[9].

A frequent approach among researchers is the use of methods based on regression [10]-[11]. In such models, in the majority of cases, the effect of selected cycling parameters on LIC degradation is analysed. Zhang *et al.* [12] developed a model which enables prediction of the remaining useful life of a battery (RUL) for a $LiNiCoAlO_2$ cell. The tests were conducted under conditions of accelerated aging – at discharge currents $I_d = 1C$ and $I_d = 2C$ and at ambient temperatures

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$T_a = 25^\circ\text{C}$ and $T_a = 40^\circ\text{C}$. In paper [13] Dessaint *et al.* implemented two multi-parametric models which allow for the determination of the maximum number of cycles which may be carried out by LiNiMnCoO₂ and LiFePO₄ cells, depending on the actual cycling parameters (I_d, I_{ch}, DoD, T_a).

In turn, Saha *et al.* in [14] used electrochemical impedance spectroscopy (EIS) and the Bayesian statistical approach to develop a new RUL method.

Moreover, as methods based on machine intelligence are being developed, it is possible to observe their more and more frequent use in problems related to the prediction of operational parameters of LICs such as, e.g. remaining useful life (RUL) [15]-[16] or capacity [17]. These methods are strictly based on data which are usually acquired from many-months of experimental procedures [18]-[21]. In [22], Hannan *et al.* proposed the use of a deep neural network to determine the SoC for LiNiMnCoO₂ cells. Li *et al.* [23] developed a method for estimating the capacity of LiFePO₄ cells, based on a convolutional neural network. Moreover, the use of support vector machines was proposed in papers by [24] to predict the state of health (SoH) of batteries. In the past, approaches based on sample entropy [25], fuzzy logic [26] and Rao-Blackwellization particle filter [27] have also been used to determine the RUL.

The GPR technique used in that paper had previously been used to predict the capacity fade or RUL. In [28], Howey *et al.* developed a model to predict the capacity fade of a LiCoO₂ cell under variable load and temperature conditions. On the other hand in [29] Hariharan *et al.* used the deep GPR to estimate the end of life (EoL) – the chemical composition of the cell was not specified. GPR was also used to predict battery degradation during calendar aging, and this issue was touched upon in paper by [30].

Because of the fact that, for some designers of systems powered from cells, SoH is not always a sufficient assessment indicator, other methods which enable the determination and prediction of the available power and energy in LICs have been developed recently. In [31], an attempt was made to predict the energy available in the battery for two cells - LiNiMnCoO₂ and LiFePO₄, using particle filtering. In a paper by [32], the state of the available energy at different I_d and at different T_a , taking into account degradation, was determined for a Li₄Ti₅O₁₂ cell using a circuit model.

B. KEY CONTRIBUTIONS

Although many different approaches have been developed to date in the aspect of modelling the effects of aging phenomena occurring in LICs, in particular the prediction of available energy and power, this issue has not yet been sufficiently explored. In many applications, not only is knowledge of the predicted useful energy of the LIC over its lifetime required, but also of the predicted changes in degradation trends. For this reason, the following contributions have been made in this paper:

1) A major contribution is the development of a model based on machine learning, which enables the prediction of the useful energy which a LIC is able to transfer during a

Fig. 1. Block diagram of the experimental procedure.

single duty cycle from begin of life (BoL) to end of life (EoL). The proposed model has a new structure of input parameters – consideration of the past energy throughput of LICs as a model parameter is an improvement of the currently existing methods.

2) The result of this research is a new dataset that can be used to test current methods and develop new predictive methods for the useful energy or SoH of LICs.

3) A demonstration of the variability of RUE_c depending on the combination of cycling parameters, and also identification of three types of evolution of changes in RUE_c in the period of operation of the LIC under consideration.

4) For the first time, the following techniques have been used: explainable artificial intelligence (XAI) – variable importance (VI) – to determine the significance of parameters of the GPR model, and accumulated local effects (ALE) – to determine their quantitative effect on RUE_c .

C. STRUCTURE OF THE PAPER

The remaining part of the paper is organised as follows. Chapter II outlines the procedure for obtaining experimental data and determining the useful energy RUE_c and also describes the results of preliminary analyses and calculations. Chapter III presents the applied methodology, GPR and XAI techniques. The results obtained and the discussion of the verification procedure are included in chapter IV.

II. DATA PREPARATION

In order to obtain data related to the ability of LICs to transfer energy during cycling, they were subjected to aging tests under various load and temperature conditions. Commercial Samsung 18650 LICs (cylindrical), with a nominal capacity of 2600 mAh and nominal voltage of 3.63 V were selected for the tests. The cathode material of the tested

TABLE I
VALUES OF CYCLING PARAMETERS SELECTED FOR AGING PROCEDURES AND RESULTS OBTAINED FROM INITIAL CALCULATIONS

Cell ID	T_c (C)	I_d (A)	I_{ch_ave} (A)	DoD (%)	FEC_{tot} (-)	RUE_c at BoL (-)	RUE_c at EoL (-)	ΔRUE_c (%)	E_{ch_tot} (Wh)
H5	18.9	-2.6	2.08	100	83	0.872	0.687	18.5	2302
A31	29	-7.8	2.62	100	184	0.827	0.418	40.9	3451
D11	17.8	-2.6	2.47	50	183	0.545	0.545	0	3435
B37	20.9	-2.6	2.35	100	72	0.872	0.709	16.3	1516
A30	17.8	-2.6	2.4	73	149	0.776	0.663	11.3	2753
H2	16.6	-2.6	1.8	27	275	0.304	0.304	0	5029
A36	25.1	-5.2	2.09	50	426	0.536	0.536	0	8002
D14	23.9	-2.6	2.15	100	130	0.88	0.74	14	2517
B40	30.3	-5.2	2.2	100	1054	0.875	0.55	32.5	19804
A35	32.5	-7.8	2.52	100	664	0.865	0.33	53.5	12449
A20	25.7	-5.2	1.81	100	209	0.942	0.704	23.8	3933
C33	32.7	-2.6	3.04	74	306	0.789	0.749	4	5765
H10	33	-2.6	2.78	74	344	0.795	0.759	3.6	6478
H9	35.3	-2.6	2.85	100	226	0.945	0.752	19.3	4254
D16	43.2	-7.8	2.64	100	905	0.911	0.294	61.7	16978
B32	39.7	-5.2	2.66	100	676	0.922	0.672	25	13573
D1	33.2	-5.2	2.38	16	3984	0.165	0.165	0	74901
A24	36.6	-5.2	2.18	50	1721	0.546	0.546	0	32340
D4	29.2	-2.6	1.2	100	1908	0.982	0.786	19.6	36109
C26	36.4	-5.2	1.9	100	909	0.97	0.689	28.1	17083
A23	36.6	-7.8	1.74	100	557	0.939	0.221	71.8	10458
H12	58.6	-7.8	2.68	100	587	0.931	0.533	39.8	10993
H1	53.8	-7.8	1.88	100	563	1.005	0.504	50.1	10555
D6	50.1	-2.6	3.46	100	375	1.046	0.813	23.3	7048
B35	47.5	-2.6	2.37	100	702	1.057	0.848	20.9	13206
A38	52.5	-5.2	2.54	100	960	1.002	0.717	28.5	18034
H13	62.1	-7.8	3.11	75	716	0.744	0.564	18	13409
C10	32.6	-2.6	2.61	100	1460	0.995	0.798	19.7	27446
D2	33.7	-2.6	3.2	100	160	0.934	0.742	19.2	3028

cells was composed of $\text{LiNi}_{0.33}\text{Mn}_{0.33}\text{Co}_{0.33}\text{O}_2$ compound, while the anode was made of graphite.

A. EXPERIMENTAL PROCEDURE

The procedure presented in Fig. 1 was adopted for the purpose of performing aging tests. Before starting the aging procedure of each cell, their reference charge (Q_{ref}) was determined in accordance with algorithm 1.

Then, each of the cells was subjected to cycling under unique temperature and load conditions. During the tests, a Panasonic MIR-254 temperature chamber was used to maintain a constant ambient temperature. Momentary temperatures of cells were recorded with probes located in central points of the cells. The average charge current from all half-cycles completed from BoL ($SoH = 100\%$) to EoL ($SoH = 80\%$) was adopted as the value of charge current during analyses. During the experimental procedures, LICs were kept in the voltage operating window specified by the manufacturer, i.e. from 2.75 V (U_{dsch}) to 4.2 V (U_{ch}). Several DoD levels were selected ranging between 16% and 100%, with an assumption that DoD is the charge which can be obtained in the given combination of cycling parameters. For example $DoD = 100\%$ corresponds to a discharge in the full voltage operating window. The cell cycling process was controlled by a Cadex C8000 tester dedicated to electrochemical cells (values of momentary currents, voltages and temperatures of LICs were measured using separate wires). To sum up, 29 cells in total were tested at 4 different T_a (10°C, 15°C, 25°C and 40°C), at 3 different values of I_d (-2.6 A, -5.2 A and -7.8 A) and at 5 different DoD levels (16%, 27%, 50%, 75% and 100%). The average value of charge current (I_{ch_ave}) ranged between 1.2 A

and 3.46 A. Table I contains detailed values of the cycling parameters adopted during the aging tests.

The condition for completion of each aging procedure was the degradation of the cell to $SoH = 80\%$. The actual SoH of the cells was checked every 48 hours under reference conditions (described in algorithm 1) using the following relationship (1)

$$SoH = \frac{Q_{ref}}{Q_{BoL}} \cdot 100\% \quad (1)$$

where Q_{ref} is the collected charge, Q_{BoL} is the charge collected before the start of the LIC cycling.

B. USEFUL ENERGY DETERMINATION

In order to determine the useful energy throughput of a LIC during a single duty cycle over its entire life, the following methodology was adopted:

1) The assumption that one complete duty cycle consists of a discharge half-cycle and the immediately following charge half-cycle.

2) Calculation of the charge throughput during a single duty cycle (collected during discharge Q_{dsch} and charge Q_{ch}) using the Coulomb Counting method, on the basis of equation (2).

Determination of the relative useful energy RUE_c throughput during the duty cycle in accordance with relationship (3). $RUE_c = 1$ means that a cell transferred nominal energy (18.876 Wh) during a full duty cycle (a discharge followed by a charge).

Algorithm 1: Q_{ref} determination

1. Interruption of the currently running aging variant after completion of the charge half-cycle (not applicable to BoL)
2. Set the temperature of the cell to 25°C
3. Charging of the cell to 4.2 V with 1.3 A current in accordance with the CC-CV protocol until the current in the CV phase drops to 0.0065 A
4. 15-second break
5. Discharge of the cell to 2.75 V with the -2.6 A current (CC method)
6. Calculation of the cell capacity using the Coulomb counting method - equation (2)

3) After each control procedure, determination of the number of completed full equivalent duty cycles ($FECs$) for the needs of the prediction model.

4) Determination of FEC_{tot} as the sum of RUE_c from all completed LIC duty cycles from BoL to EoL. An assumption was made that the transfer of nominal energy (18.876 Wh) during two consecutive charge and discharge half-cycles constitutes a full equivalent cycle FEC .

5) Determination of the total energy throughput $E_{th,tot}$ as a product of total charge throughput of LIC, multiplied by its voltage on terminals.

$$Q_{ch/dsch} = Q_0 + \int_0^t I_{ch/dsch}(t) dt \quad (2)$$

$$RUE_c = \frac{Q_{dsch} \cdot U_{ave\_dsch} + Q_{ch} \cdot U_{ave\_ch}}{2 \cdot C_{nom} \cdot U_{nom}} \quad (3)$$

where Q_0 is the initial charge accumulated in a LIC, U_{ave_dsch} is an average voltage during a discharge half-cycle, U_{ave_ch} is an average voltage during a charge half-cycle, C_{nom} is the nominal capacity and U_{nom} is the nominal voltage.

C. INITIAL ANALYSIS AND CALCULATIONS

The tests carried out showed that for the LICs discharged to $DoD > 70\%$ the value of the transferred RUE_c is variable during their considered life. In the case of variants with $DoD < 70\%$, LICs transferred the fixed energy throughout their life ($RUE_c = \text{const}$). Depending on the combination of the values of the cycling parameters, three types of evolution of RUE_c changes were observed during the analysed period of operation: a) approximately linear, b) a slow decrease in the first phase of life followed by a phase of accelerated loss of

energy transfer, c) a characteristic inflection point occurred after the phase of releasing the assumed charge, followed by a phase of rapid loss of energy transfer. The phenomena described in points b) and c) were present in cells "A23", "D14", "D16", "A30", "H10" and "C33". The selected characteristics of the RUE_c of cells, in which the effect described above occurred, are presented in Fig. 2, and the results of the remaining experimental measurements of the cells, are available online (raw data: <https://data.mendeley.com/datasets/fzp5wx28kw/1>).

For each completed aging variant, it was checked how the ability of the cell to transfer RUE_c decreased. For this purpose, the ΔRUE_c difference (eq. 4) between RUE_c determined at

Fig. 2. Three types of evolution of RUE_c trends observed for the tested LIC. BoL and EoL was calculated.

$$\Delta RUE_c = (RUE_{c\_BoL} - RUE_{c\_EoL}) \cdot 100\% \quad (4)$$

Depending on the conditions under which the LICs were cycled, ΔRUE_c reached values exceeding 50% (cells: "A35", "D16", "H1" and "H23") which means that in the period preceding EoL the cells were not capable of transferring even half of the initial RUE_c . A high value of ΔRUE_c was observed for cells discharged with a high current (-7.8 A) regardless of the temperature conditions (average cyclic temperature of the cell). The lowest values of ΔRUE_c occurred for cycled cells at incomplete DoD ("A30", "C33", "H10" and "H13"). Additionally, FEC_{tot} and $E_{th,tot}$ completed by each LIC were determined and listed for information purposes in Table I. Results of initial calculations are included in Table I.

III. PROPOSED METHODOLOGY

A. PROPOSED MODEL

The amount of RUE_c which LICs are capable of transferring during a full duty cycle over their lifetime depends on the combination of values of cyclic operating parameters and the number of completed $FECs$. As the tests demonstrated, LICs may have a similar ability to transfer RUE_c even if they have completed a significantly different number of $FECs$. One additional issue is the fact that the cyclic operating parameters of a LIC are strongly correlated. For instance higher values of DoD , I_d and I_{ch_avg} result in an increase in the average temperature of the LIC, which, in turn, determines the amount of RUE_c . Furthermore, the impossibility of separating the impact of the above-mentioned parameters during a duty cycle makes it highly difficult to determine the precise effect of individual parameters on the amount of RUE_c . In this paper, in order to develop a prediction model for the relative useful energy RUE_c the framework presented in Fig. 3 was adopted. Taking the above-mentioned challenges into account, the author proposes the use of the non-parametric model, whose structure strictly matches the form of learning data. This model belongs to the group of machine learning models – it is more frequently referred to as GPR and is a non-parametric regression technique based on Gauss processes. The use of a non-parametric model forces out the use of techniques which enable the interpretation of the manner in which the implemented non-parametric model operations.

Fig 3. General flowchart of the applied framework.

B. GAUSSIAN PROCESS REGRESSION

Gaussian process regression (GPR) belongs to the group of kernel-based non-parametric models [33]-[34]. In this model, the Gaussian probability distribution is defined for each finite set of input variables x_i .

$$f(x) \sim GP(\mu(x), \text{cov}(x, x')) \quad (5)$$

where $\mu(x)$ is mean function, and $\text{cov}(x, x')$ is the covariance function (kernel function).

The kernel functions contain parameters related to the scaling of responses of model y and input vector x and are referred to as hyperparameters θ . For the majority of standard kernels, the hyperparameters include: standard deviation σ_f and characteristic length σ_l .

The matching of the structure of the model to the learning set takes place by defining, for each sample of the learning dataset x_i , a function $f(x_i)$ having the Gaussian distribution – hereafter called a hidden variable – and a set of basis functions $h(x)$, which transfer the input vector x_i from the original feature space R^d to the extended feature space R^p .

For the needs of the RUE_c prediction, the following structure of the model has been adopted

$$y(x) = h(x)^T w + f(x) + \varepsilon \quad (6)$$

where $h(x)$ is the set of basis functions, w is the vector of basis function coefficients, ε is noise with normal distribution.

Model prediction y takes place on the basis of input vector x' and the learning dataset

$$P(y_i | f(x_i), x_i) \sim N(y_i | h(x_i)^T w + f(x_i), \sigma^2) \quad (7)$$

C. VARIABLE IMPORTANCE

Variable importance is the technique which allows for the determination of the significance of each explanatory variable in reference to its effect on the implemented machine learning model [35]-[36]. This concept consists of the determination of the model error by permutations of the values of each variable. The given variable is significant if changes in its value cause changes in the model error, as the model relied on this variable

Algorithm 2: Variable importance determination

Input:

X – variables matrix (explanatory variables)
 y – vector of predicted values of original data
 f – trained model
 $L(y, f)$ – loss function (any model performance method)

Steps:

1. Compute the original model error $L^0 = L(y, f(X))$
2. For each explanatory variable X_j :
  - a. In order to break the relationship between variable X_j and true prediction y – generate feature matrix X^p by permuting variable X_j in the data X .
  - b. Compute model predictions Y based on the permuted data
  - c. Calculate error based on the predictions of the permuted data $L_j = L(Y, f(X^p))$.
  - d. Calculate variable importance $VI_j = L_j - L^0$
3. Sort explanatory variables by descending VI

Output:

VI_j – variable importance of each explanatory variable

for prediction. If the permutations of the variable do not cause changes in the model error, then it is considered insignificant.

In other words, the more the model relies on a variable to make predictions, the more important it is for the model. The values of significance of the variables are determined on the basis of algorithm 2.

D. ACCUMULATED LOCAL EFFECTS

Accumulated local effects (ALE) is the method which allows for the assessment of the relationship between the explanatory variables of the model and the predicted value [36]-[37]. ALE reflects the way in which model forecasts change within the narrow range of values of variables. This method is particularly useful in the case of sets whose parameters are correlated with each other. The ALE function is defined as follows:

$$ALE(x_1) = \int_{\min(x_1)}^{x_1} E \left[\frac{\partial f(x_1, x_2)}{\partial x_1} \middle| x_1 = z_1 \right] dz_1 + c_1 = \quad (8)$$

where partial derivative $\frac{\partial f(x_1, x_2)}{\partial x_1}$ represents the local effect of x_1 on prediction function f at (x_1, x_2) , $\min(x_1)$ is lower value of considered variable range, c_1 is a constant selected in order to vertically center the graph.

Averaging the local effect over the conditional distribution $p(x_2, x_1)$ makes it possible to separate the effect of other correlated variables located outside the considered range of values of variable x_1 .

In order to determine local effects, the whole range of values of the given explanatory variable must be divided into multiple intervals. The "effect" is understood as the difference in prediction calculated separately for each occurrence of a variable in a given interval. All differences in the given interval are summed up and divided by the number of occurrences – this way, the obtained mean difference in predictions is the "local" effect. Ultimately the value of the

Fig 4. Learning results obtained for three observed types of RUE_c evolution. (a) After about 300 $FECs$, the cell lost the ability to transfer the fixed energy – the phase of significant decrease in RUE_c takes place ($MAPE=0.025\%$). (b) Under these conditions, throughout the lifetime of the LIC, the decrease in RUE_c was approximately linear ($MAPE=0.003\%$). (c) The phase of slow degradation occurs until approximately 400 $FECs$ are completed, then it is possible to observe the phase of accelerated ability to transfer RUE_c ($MAPE=0.176\%$).

accumulated local effect of a given variable is determined by summing up all the local effects and represents the average change in the prediction when the value of the variable changes within its range.

The ALE method also allows for analysis of changes in the predicted value in the case of interaction of two (ALE second-order) or more variables (ALE higher-order). In this case, the main impact of the variables concerned is ignored – only the effect of their interaction is calculated. For two explanatory variables, the ALE function may be represented as

$$ALE(x_1, x_2) = \int_{\min(x_1)}^{x_1} \int_{\min(x_2)}^{x_2} E \left[\frac{\partial f(x_1, x_2, x_{(1,2)})}{\partial x_1 \partial x_2} \right]_{x_1 = z_1, x_2 = z_2} dz_1 dz_2 \quad (9)$$

The rules for the determination of ALE for two variables are identical to those for one variable, whereby, the intervals are replaced by rectangular areas due to the necessity of accumulation of local effects in two dimensions.

IV. RESULTS AND DISCUSSION

The input parameters of the model were the cyclic operating parameters: the average LIC temperature during the full duty cycle (T_c), the value of current during the discharge half-cycle (I_d), depth of discharge (DoD), the value of mean current during the charge half-cycle ($I_{ch,avg}$) and the number of full equivalent cycles ($FECs$). The predicted value was the relative useful energy (RUE_c) throughput of a cell during a full duty cycle.

A. MODEL TRAINING

Before initiating the machine learning procedure, the data obtained from the experimental measurements were normalised. After the normalisation, the average value of each of the parameters of the learning dataset was equal to 0, and the standard deviation was equal to 1. The results of measurements of cycling of LICs, which were cycled with $DoD > 70\%$ (20 LICs in total), were used as the learning dataset of the non-parametric GPR model. Cells "A38", "B32", "B40" and "H9" were used to verify the model exclusively. Finally, the learning dataset contained 311 samples, and the verification set consisted of 99 samples. The value of $FEC=0$ is equivalent to $SoH=100\%$ of the cell, while the last value of $FEC=533$ indicates $SoH=80\%$ and the

TABLE II
RESULTS OF LEARNING PROCEDURE AND VALUES OF
CHARACTERISTIC KERNEL PARAMETERS

	σ_l (-)	σ_f (-)	MAPE (%)	R^2 (-)	L (-)	RMSE (-)
GPR model	0.7659	0.1687	0.05	0.99	713.7	0.014144

TABLE III
RESULTS OF VERIFICATION PROCEDURE ON NEW LOAD PROFILES

Cell ID	T_c (°C)	I_d (A)	DoD (%)	$I_{ch,avg}$ (A)	MAPE (%)
B40	30.3	-5.2	100	2.2	5.63
H9	35.3	-2.6	100	2.85	1.05
A38	54.6	-5.2	100	2.63	2.77
B32	39.7	-5.2	100	2.47	3.26

accomplishment of the EoL status. As the kernel function of the GPR model, the matern 3/2 function was used, which is expressed by the following equation:

$$k_{mat3/2}(x, x') = \sigma_f^2 \left(1 + \frac{\sqrt{3}r}{\sigma_l} \right) \exp \left(-\frac{\sqrt{3}r}{\sigma_l} \right) \quad (10)$$

where r is Euclidean distance between x and x' .

The values of characteristic parameters σ_f and σ_l were determined using the method of optimisation without constraints (quasi-Newton).

Linear function type was adopted for the function set $h(x)$. The coefficient of determination R^2 , mean average percentage error $MAPE$ and root mean square error ($RMSE$) were used to assess the model quality.

The results of the learning process of the GPR model: the values of the respective characteristic parameters are listed in Table II, while the example characteristics of the three observed types of RUE_c evolution are presented in Fig. 4 (a-c). The learning results of other LICs are included in Appendix A. For each LIC included in the learning dataset, the response of the model was within the adopted confidence interval (95%). The $MAPE$ of the model developed for all samples of the learning dataset was 0.05%. This proves that the GPR model almost perfectly matches the learning dataset.

B. PREDICTION

The developed original model was verified using new load profiles which were not used to train the model. LICs with different mean cycling temperatures (T_c) at mean load current of $I_d = -5.2$ A were selected for verification. For instance, for $T_c > 40^\circ\text{C}$, the learning dataset included LICs loaded with

Fig. 5. Results of the model verification procedure.

$I_d = -2.6$ A and $I_d = -7.8$ A (cells "D6" and "H12"), therefore, the "A38" cell was selected for verification at higher temperatures. The verification process consisted of the RUE_c prediction by a model of 4 cells in the period between BoL and EoL , and of comparison of their values obtained during the experiment. The verification results are presented in Table III, and the characteristics for the 4 tested cases are presented in Fig. 5.

In the author's opinion, the obtained results of the verification process confirm the effectiveness of the selected GPR method for the purpose of RUE_c prediction which a LIC can transfer during a full duty cycle throughout its life. Only in one test case did the $MAPE$ slightly exceed 5% (the "B40" cell). Also in favour of the method used, is the fact that despite an almost ideal matching of the model to the learning data, the prediction of RUE_c under new operating conditions for the LIC is possible with little error. The GPR model also allows for prediction within ranges exceeding the ranges of variables of the learning dataset, which is not possible for some techniques based on artificial intelligence – for instance, neural networks or fuzzy logic.

C. MODEL EXPLANATION

In order to determine the significance of the explanatory variables of the model, the variable importance (VI) technique was used. $RMSE$ was selected as the loss function of L model. The calculations were repeated 8 times in order to negate the effect of uncertainty caused by permutations of values. The obtained results of VI are characterised by a low standard deviation; they were averaged and listed in Table IV. Based on the results, the respective model input parameters were ordered in terms of their significance - the effect of FEC on the predicted value of RUE_c is the highest in the used dataset (the highest VI), then the effect of I_d and T_c is on a comparable level. From among the adopted model parameters, DoD and I_{ch_avg} have the lowest effect on RUE_c (in both cases the calculated significance is at a similar level).

Given the fact that interpretation of the structure of the developed GPR non-parametric model is very difficult, in order to determine the effect which the respective model parameters have on the value of the RUE_c prediction, the author has applied the ALE concept. Fig. 6 (a–e) presents the plots of the first-order accumulated local effects which determine the impact of individual input parameters of the GPR model on RUE_c . The value of ALE at the given point determines the difference in prediction in relation to the mean prediction of RUE_c . $ALE=0$ represents the mean prediction of RUE_c . For instance, for $T_c \approx 25^\circ\text{C}$, the RUE_c prediction will be lower by 0.05 in relation to the mean prediction which is an

TABLE IV
VALUES OF VARIABLES IMPORTANCE OF IMPLEMENTED MODEL

Parameter	T_c ($^\circ\text{C}$)	I_d (A)	DoD (%)	I_{ch_avg} (A)	FEC (-)
mean VI	0.120042	0.147175	0.048705	0.056989	0.167583
std. dev.	0.001375	0.001651	0.000866	0.000554	0.001614

Fig. 6. First-order ALE plots for individual model input parameters (a) average temperature of LIC during the full cycle (b) discharge current (c) depth of discharge (d) average charge current (e) full equivalent cycle.

exclusive consequence of temperature (the effect of other cyclic operating parameters is separated). Based on the results obtained, it is possible to determine, unambiguously, the effect of T_c on RUE_c – the lowest values of RUE_c are obtained at average cycling temperatures below 30°C and they increase gradually up to the range of values between 30°C and 44°C , where RUE_c reaches its peak value and then falls for T_c higher than 44°C (Fig. 6 – a). In the case of values of currents I_d and I_{ch_avg} , the effect on RUE_c is linear. The lower the values of I_d and I_{ch_avg} are, the higher the RUE_c is. Furthermore, cycling of the LICs at I_d values lower than -5.2 A and higher than 2.25 A for I_{ch_avg} will result in lower RUE_c values over their lifetime – the visible breaking point of characteristics (Fig. 6 – b, d). The plot of ALE for DoD showed the linear dependence of this factor on RUE_c (Fig. 6 – c). The amount of transferred RUE_c drops sharply when the cells complete more than 500 full equivalent duty

Fig. 7. Plots of accumulated local effects of the second order for the respective combinations of pairs of model input parameters. cycles (Fig. 6 – e).

Additionally, the results obtained using the ALE method confirm the results obtained using the VI technique – the highest dispersion of ALE values (from -0.286 to 0.225) is characteristic of FEC , which is compliant with the highest value of VI for this parameter. For I_d and T_c the differences in relation to the mean prediction vary from -0.114 to 0.08 for I_d and from -0.108 to 0.027 for T_c respectively, which also correlates with the VI results. The lowest dispersion of values was observed for I_{ch_avg} – from -0.063 to 0.031, which means the lowest effect on RUE_c in the used dataset.

In order to determine the effect of interaction of the model parameters on RUE_c , the values of the second-order accumulated local effects were determined in the paper for all the possible combinations of pairs of the GPR model input parameters. The results obtained in the form of value maps are presented in Fig. 7 (a–j). Analysis of the interaction effect of model parameter pairs showed that the highest influence on RUE_c is exerted by combinations of the following pairs $T_c - FEC$, $I_d - FEC$ and $I_{ch\_avg} - FEC$. For instance, for LICs, which completed a large number of FEC s (over 1500) at T_c below 25°C, the differences in relation to the mean RUE_c prediction even amount to -0.4, and at T_c above 50°C +0.2 respectively (Fig. 7 a). A high level of influence was also shown by the $I_{ch\_avg} - FEC$ pair at around $FEC=1350$ and $I_{ch\_avg}=3A$ – the ALE value in this area reaches up to +0.3 (Fig. 7 d). The level of influence of the $DoD - FEC$ pair over the entire value range does not exceed the range between -0.04 and +0.06 (Fig. 7 d). To sum up – the pairs containing FEC are characterised by the highest influence due to the highest effect of FEC on RUE_c which is shown in Fig. 6 – e. The interaction effect of parameter pairs, i.e. $T_c - DoD$ and $T_c - I_d$ is locally at the level of hundredths (+0.02 and -0.015) – Fig. 7 – h,j. On the other hand, the effect of other pairs is at a negligible level (ALE at a level of thousandths or lower) – Fig. 7 – e-g,i.

APPENDIX

A. RESULTS OF THE LEARNING PROCEDURE

Cell ID	MAPE (%)	Cell ID	MAPE (%)	Cell ID	MAPE (%)
D14	0.00424	B35	0.036	C26	0.0088
A35	0.1015	A30	0.1034	D2	0.0149
A20	0.0219	B37	0.1902	A25	0.0393
D16	0.0262	A31	0.2897	D6	0.0095
C10	0.0009	H5	0.232	C33	0.02779
A23	0.1761	D4	0.0031	H12	0.00789
H10	0.02456	H1	0.03494		

REFERENCES

- [1] R. Pietrachio *et al.*, “Potential of Using Medium Electric Vehicle Fleet in a Commercial Enterprise Transport in Germany on the Basis of Real-World GPS Data,” *Energies*, vol. 14(17), pp. 1–23, 2021.
- [2] K. Edström *et al.*, “BATTERY 2030+ Roadmap,” p. 83, 2020.
- [3] Z. Chen, Y. Ren, A. N. Jansen, C. K. Lin, W. Weng, and K. Amine, “New class of nonaqueous electrolytes for long-life and safe lithium-ion batteries,” *Nat. Commun.*, vol. 4, pp. 1–8, 2013.
- [4] M. Broussely *et al.*, “Main aging mechanisms in Li ion batteries,” *J. Power Sources*, vol. 146, no. 1–2, pp. 90–96, 2005.
- [5] J. Vetter *et al.*, “aging mechanisms in lithium-ion batteries,” *J. Power Sources*, vol. 147, no. 1–2, pp. 269–281, 2005.
- [6] A. Wang, S. Kadam, H. Li, S. Shi, and Y. Qi, “Review on modeling of the anode solid electrolyte interphase (SEI) for lithium-ion batteries,” *npj Comput. Mater.*, vol. 4, no. 1, 2018.
- [7] J. Olmos, I. Gandiaga, A. Saez-de-Ibarra, X. Larrea, T. Nieva, and I. Aizpuru, “Modelling the cycling degradation of Li-ion batteries: Chemistry influenced stress factors,” *J. Energy Storage*, vol. 40, no. May, p. 102765, 2021.
- [8] M. Ecker *et al.*, “Development of a lifetime prediction model for lithium-ion batteries based on extended accelerated aging test data,” *J. Power Sources*, vol. 215, pp. 248–257, 2012.
- [9] W. Diao, S. Saxena, and M. Pecht, “Accelerated cycle life testing and capacity degradation modeling of LiCoO₂-graphite cells,” *J. Power Sources*, vol. 435, no. June, p. 226830, 2019.
- [10] M. Naumann, F. Spingler, and A. Jossen, “Analysis and modeling of cycle aging of a commercial LiFePO₄/graphite cell,” *J. Power Sources*, vol. 451, no. December 2019, p. 227666, 2020.

- [11] L. Tao, J. Ma, Y. Cheng, A. Noktehdan, J. Chong, and C. Lu, "A review of stochastic battery models and health management," *Renew. Sustain. Energy Rev.*, vol. 80, no. February 2018, pp. 716–732, 2017.
- [12] R. Xiong, Y. Zhang, J. Wang, H. He, S. Peng, and M. Pecht, "Lithium-Ion Battery Health Prognosis Based on a Real Battery Management System Used in Electric Vehicles," *IEEE Trans. Veh. Technol.*, vol. 68, no. 5, pp. 4110–4121, 2019.
- [13] S. N. Motapon, E. Lachance, L.-A. Dessaint, and K. Al-Haddad, "A Generic Cycle Life Model for Lithium-Ion Batteries Based on Fatigue Theory and Equivalent Cycle Counting," *IEEE Open J. Ind. Electron. Soc.*, vol. 1, no. August, pp. 207–217, 2020.
- [14] B. Saha, K. Goebel, S. Poll, and J. Christophersen, "Prognostics methods for battery health monitoring using a Bayesian framework," *IEEE Trans. Instrum. Meas.*, vol. 58, no. 2, pp. 291–296, 2009.
- [15] K. Liu, Y. Shang, Q. Ouyang, and W. D. Widanage, "A Data-Driven Approach with Uncertainty Quantification for Predicting Future Capacities and Remaining Useful Life of Lithium-ion Battery," *IEEE Trans. Ind. Electron.*, vol. 68, no. 4, pp. 3170–3180, 2021.
- [16] S. A. Hasibet *et al.*, "A Comprehensive Review of Available Battery Datasets, RUL Prediction Approaches, and Advanced Battery Management," *IEEE Access*, vol. 9, pp. 86166–86193, 2021.
- [17] R. R. Richardson, C. R. Birkl, M. A. Osborne, and D. A. Howey, "Gaussian Process Regression for in Situ Capacity Estimation of Lithium-Ion Batteries," *IEEE Trans. Ind. Informatics*, vol. 15, no. 1, pp. 127–138, 2019.
- [18] G. dos Reis, C. Strange, M. Yadav, and S. Li, "Lithium-ion battery data and where to find it," *Energy AI*, vol. 5, no. May, 2021.
- [19] C. Vidal, P. Malysz, P. Kollmeyer, and A. Emadi, "Machine Learning Applied to Electrified Vehicle Battery State of Charge and State of Health Estimation: State-of-the-Art," *IEEE Access*, vol. 8, pp. 52796–52814, 2020.
- [20] D. Burzyński and L. Kasprzyk, "A novel method for the modeling of the state of health of lithium-ion cells using machine learning for practical applications," *Knowledge-Based Syst.*, vol. 219, p. 106900, 2021.
- [21] L. Kasprzyk, A. Domeracka, D. Burzyński, "Modelowanie pracy i trwałości akumulatorów litowo-jonowych w pojazdach elektrycznych," *Przegląd Elektrotechniczny*, no. 12, 160–163, 2018.
- [22] D. N. T. How, M. A. Hannan, M. S. H. Lipu, K. S. M. Sahari, P. J. Ker, and K. M. Muttaqi, "State-of-Charge Estimation of Li-Ion Battery in Electric Vehicles: A Deep Neural Network Approach," *IEEE Trans. Ind. Appl.*, vol. 56, no. 5, pp. 5565–5574, 2020.
- [23] Y. Li, K. Li, X. Liu, and L. Zhang, "Fast battery capacity estimation using convolutional neural networks," *Trans. Inst. Meas. Control*, 2020.
- [24] I. H. Li, W. Y. Wang, S. F. Su, and Y. S. Lee, "A merged fuzzy neural network and its applications in battery state-of-charge estimation," *IEEE Trans. Energy Convers.*, vol. 22, no. 3, pp. 697–708, 2007.
- [25] X. Hu, J. Jiang, D. Cao, and B. Egardt, "Battery health prognosis for electric vehicles using sample entropy and sparse Bayesian predictive modeling," *IEEE Trans. Ind. Electron.*, vol. 63, no. 4, pp. 2645–2656, 2016.
- [26] D. Burzyński, R. Pietrach, L. Kasprzyk, and A. Tomczewski, "Analysis and Modeling of the Wear-Out Process of a Lithium-Nickel-Manganese-Cobalt Cell during Cycling Operation under Constant Load Conditions," *Energies*, vol. 12, no. 20, p. 3899, Oct. 2019.
- [27] A. El Mejdoubi, H. Chaoui, H. Gualous, P. Van Den Bossche, N. Omar, and J. Van Mierlo, "Lithium-ion batteries health prognosis considering aging conditions," *IEEE Trans. Power Electron.*, vol. 34, no. 7, pp. 6834–6844, 2019.
- [28] R. R. Richardson, M. A. Osborne, and D. A. Howey, "Battery health prediction under generalized conditions using a Gaussian process transition model," *J. Energy Storage*, vol. 23, no. March, pp. 320–328, 2019.
- [29] P. Tagade *et al.*, "Deep Gaussian process regression for lithium-ion battery health prognosis and degradation mode diagnosis," *J. Power Sources*, vol. 445, no. October 2019, p. 227281, 2020.
- [30] K. Liu, Y. Li, X. Hu, M. Lucu, and W. D. Widanage, "Gaussian Process Regression with Automatic Relevance Determination Kernel for Calendar Aging Prediction of Lithium-Ion Batteries," *IEEE Trans. Ind. Informatics*, vol. 16, no. 6, pp. 3767–3777, 2020.
- [31] R. Xiong, Y. Zhang, H. He, X. Zhou, and M. G. Pecht, "A double-scale, particle-filtering, energy state prediction algorithm for lithium-ion batteries," *IEEE Trans. Ind. Electron.*, vol. 65, no. 2, pp. 1526–1538, 2017.
- [32] K. Li, F. Wei, K. J. Tseng, and B. H. Soong, "A Practical Lithium-Ion Battery Model for State of Energy and Voltage Responses Prediction Incorporating Temperature and aging Effects," *IEEE Trans. Ind. Electron.*, vol. 65, no. 8, pp. 6696–6708, 2018.
- [33] C. E. Rasmussen and Ch. K. Williams, "Gaussian processes for machine learning," *MIT press Cambridge*, vol. 1, 2006.
- [34] S. Haykin and S. Simon, "Neural Networks and learning machines," *Prentice Hall*, New York, 2009.
- [35] P. Wei, Z. Lu, and J. Song, "Variable importance analysis: A comprehensive review," *Reliab. Eng. Syst. Saf.*, vol. 142, pp. 399–432, 2015.
- [36] C. Molnar, "Interpretable Machine Learning. A Guide for Making Black Box Models Explainable," *Book*, p. 247, 2019.
- [37] D. W. Apley and J. Zhu, "Visualizing the effects of predictor variables in black box supervised learning models," *J. R. Stat. Soc. Ser. B Stat. Methodol.*, vol. 82, no. 4, pp. 1059–1086, 2020.

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