A grey-box approach for the Prognostic and Health Management of Lithium-Ion Batteries

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ABSTRACT

The lithium-ion batteries (LIB) industry is rapidly growing and is expected to continue expanding exponentially in the next decade. LIBs are already widely used in everyday life, and their demand is expected to increase further, particularly in the automotive sector. The European Union has introduced a new law to ban internal combustion engines from 2035, pushing for the adoption of electric vehicles and increasing the need for more efficient and reliable energy storage solutions such as LIBs. As a result, the establishment of Gigafactories in Europe and the United States is accelerating to meet the growing demand and partially reduce dependencies on China, which is currently the main producer of LIBs.

To fully realize the potential of LIBs and ensure their safe and sustainable use, it is crucial to optimize their useful life and develop reliable and robust methodologies for estimating their state of health and predicting their remaining useful life. This requires a comprehensive understanding of LIB behaviour and the development of effective prognostic and health management approaches that can accurately predict battery degradation, plan for maintenance and replacements, and improve battery performance and lifespan.

This work, funded by the GREYDIENT project, a European consortium aiming to advance the state of the art in the grey-box approach, combines physical modeling (white box) and machine learning (black box) techniques to demonstrate the grey-box effectiveness in the prognostic and health management. The grey-box approach here proposed consists of a combination of a physical battery model whose degradation parameters are estimated online at every cycle by a multi-layer perceptron particle filter (MLP-PF).

An electrochemical degradation model of a Lithium-Ion battery cell has been derived by use of Modelica. The model simulates the output voltage of the cell, while the degradation over time is simulated through the variation of 3 parameters: q_{Max} (maximum number of lithium-ions available), R_0 (internal resistance) and D (diffusion coefficient). To validate the model we resort to the well-known NASA Battery dataset, which has also been used to infer the optimal values of the three hidden degradation parameters at every cycle, to obtain their run-to-failure history.

Then, the physical model is combined with the MLP-PF: an MLP artificial neural network is firstly trained on the run-to-failure degradation processes of the model parameters, allowing the propagation of the parameters in the future and the corresponding estimation of the battery remaining useful life (RUL). The MLP is then updated online by a particle filter every time a new measurement is available from the battery management system (BMS), providing flexibility to this method, needed for the electrochemical nature of the batteries, and allowing the propagation of uncertainties.

1. INTRODUCTION

Electric vehicles increased their market share constantly in the last 10 years, with their number tripled in the last 3 (International Energy Agency (IEA), 2022). Their success is driven by multiple factors: (i) the number of models present on the market is constantly increasing, with more than 400 EV types available in 2021, (ii) the political support, expressed through incentives and subsidies, to phase out from

the internal combustion engine (ICE) (in this regard, the European Union approved a law forbidding the selling of combustion engine vehicles starting from 2035) to contrast air pollution and climate change; (iii) the use of more efficient batteries, allowing the use of EV for comparable driving range with respect to the ICE vehicles.

Lithium-Ion Batteries (LIBs) are the preferred solution for EVs, due to their longevity and efficiency with respect to other batteries' typologies. Moreover, LIBs prices have continuously decreased during the last few years, leading to more affordable vehicles, hence contributing to their development. However, due to the shortage in the supply chain and the rising cost of metals, in 2022 the price for the battery packs increased for the first time since 2010 (BloombergNEF, 2022). Despite this, the demand for Li-Ion batteries is predicted to grow in all regions: China remains the first manufacturer, but numerous projects are aiming to exponentially increase production in Europe and the United States to loosen their dependence. The projects involve the construction of Lithium-Ion batteries gigafactories, with an expected total production of about 900 Gwh in 2030 (Heiner, Heimes, 2022).

In this context, the research of novel and more efficient methods for the optimisation of the usage of batteries is of crucial importance. The implementation of effective maintenance strategies can lead to an improvement in battery longevity, resulting in a decrease in battery operating costs. Indeed, LIB performances degrade with usage and over time. This will eventually lead to the inability to supply the required amount of power, with the need to replace the battery to guarantee the right performance for the vehicle. On the other side, if used improperly the failure can be sudden, and lead to the thermal runaway of the battery, with consequences ranging greatly, including the possible complete burning of the vehicle (Sun, Bisschop, Niu, & Huang, 2020).

To avoid this, EVs are equipped with a battery management system (BMS), which monitors and controls the parameters of the battery to maintain safe and efficient operations. The data collected by the battery sensors offer opportunities to develop diagnosis and prognosis strategies to further improve the safety and optimize and maximize the remaining useful life (RUL) of the battery. This consequently allows for deriving smarter strategies for the batteries' recharge and replacement.

Given the strategic importance of the matter, many works have been dedicated to the prognosis and diagnosis of LIBs in literature; However, given the complicated internal electrochemical mechanism that leads to strong non-linear degradation behaviour, these remain extremely complex challenges. (Hu, Xu, Lin, & Pecht, 2020) provide an overview of the methods for the prognostic and health management (PHM) of the batteries, clustered in model-based, data-driven and hybrid methods.

A model-based approach relies on the physical modelling of the degradation processes of the battery to describe its trajectory, while data-driven methods use statistical or machine learning techniques as a surrogate for the physical model. Model-based approaches allow for deeply characterised behaviour at different levels of precisions, from relatively simple equivalent circuit models, as in (Zou, Hu, Ma, & Li, 2015), to more complex complete electrochemical ones (Lyu et al., 2017). They also allow to focus on a specific phenomenon, for example in (Tran et al., 2022) various methods are reviewed, focusing on the diagnosis of the thermal runaway of the battery.

Data-driven methods use historical monitoring data to build a surrogate of the battery and predict its performance and RUL. These methods focus on the degradation trend, while completely ignoring the propagation and internal mechanism. Given their flexibility and the worldwide interest in machine learning, and in particular artificial neural networks (ANN), many works proposed this approach in the last years (Zhang, Xiong, He, & Pecht, 2018; Khaleghi et al., 2022; Wang et al., 2021). If the results are often acceptable in terms of prediction error and computational costs, concerns remain on the lack of interpretability, and on the availability of the necessary amount of historical data for the particular application.

To overcome the limitations of model-based (computational costs, expert knowledge of the phenomena involved) and data-driven (interpretability, amount of data), many hybrid approaches have been proposed. The most promising one is based on physics-informed neural network (PINN). (Li et al., 2021) focused on the estimation of the internal hidden states of the battery by combining an electrochemical-thermal model and a deep neural network, while (Shi, Rivera, & Wu, 2022) included physics in a long-short term memory (LSTM) model. Finally, in (Nascimento, Corbetta, Kulkarni, & Viana, 2021) an electrochemical model of the battery is combined with a deep neural network for uncertainty quantification to improve the agreement with experimental data.

This work proposes a different hybrid approach, consisting of the combination of a physics-based battery model in which the degradation parameters are estimated online with a data-driven approach, resulting in a so-called grey-box methodology. To do so, it is defined a framework composed of three main areas: calibration, propagation and prognostic. The first, calibration, is needed to calibrate the physical model, which evolves over time due to the electrochemical nature of the problem. The battery electrochemical degradation model has been developed based on (Daigle & Kulkarni, 2013) by means of Modelica language (OpenModelica, 2007). The model simulates the output voltage of the battery and resorts to 3 parameters (maximum number of lithium-ions available q_{Max}, internal resistance R_0 and diffusion coefficient D) to describe its degradation over time. These hid-

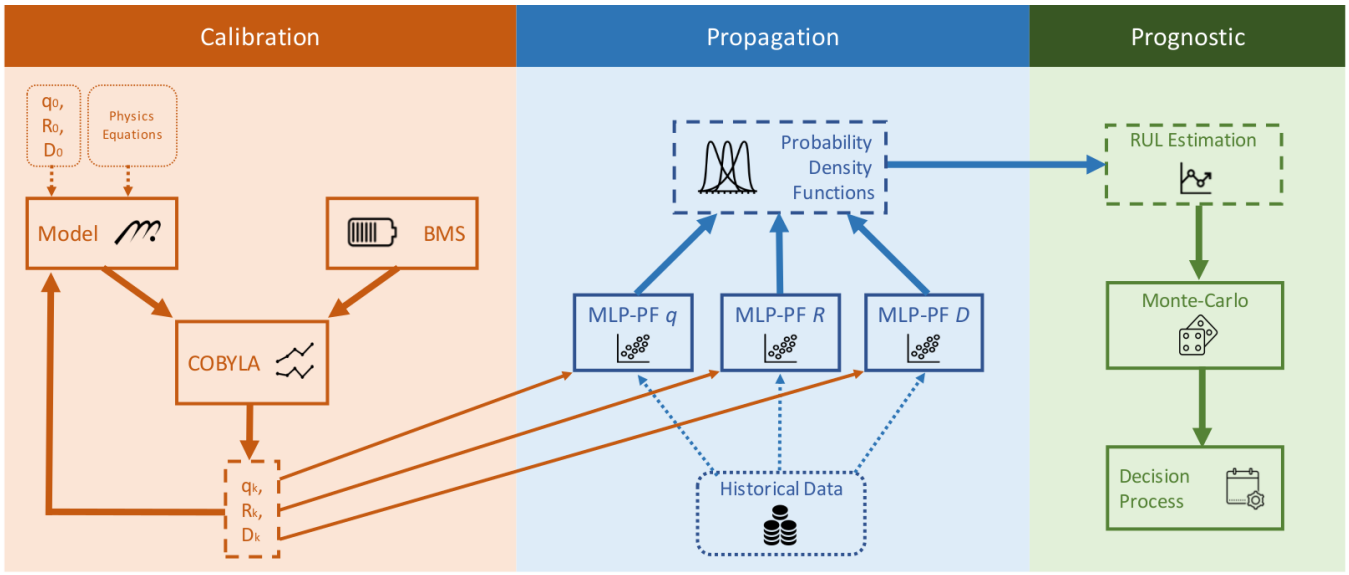

Figure 1. Illustration of the framework for the Prognostic of the battery RUL.

den model parameters are inferred and updated every time a new measurement is available from the BMS through the COBYLA (Powell, 1994) optimisation algorithm. The second area, propagation, resorts to a multi-layer perceptron particle filter (MLP-PF) (Cadini, Sbarufatti, Cancelliere, & Giglio, 2019) to propagate the three hidden parameters in the future. This methodology allows to compute the probabilities density functions of the parameters in each future instant of time, allowing the estimation of the battery RUL and the evaluation of the model in a Monte-Carlo fashion, in the prognostic phase.

This paper is organized as follows: Section 2 describes the proposed PHM framework. In Section 2.1 the model is described and the optimisation process used to infer the parameters is proposed. Then Section 2.2 introduces the concept of the MLP-PF, and the results obtained with the proposed approach are shown in Section 2.3. Finally, Section 3 derives some conclusions and future developments on this work.

2. PHM FRAMEWORK

The final goal of the proposed work is to derive an easy-to-understand indicator for the final customer to allow decision making, which can be an indication for when to replace the battery in the case of a private vehicle or planning maintenance strategies in the case of a fleet. To do so, we resort to the online framework illustrated in Figure 1. It is characterised by three main areas: calibration, propagation and prognostic.

The first area concerns the calibration of the battery during usage: due to the electrochemical properties of the LIBs, batteries with the same physical characteristics can behave and degrade very differently one from another, depending on the external conditions, usage and possible inner defects. To account for that, the model needs to be calibrated with respect to the measurement coming from the BMS: every time a measurement is available, the parameters responsible for the battery degradation are updated, allowing a better simulation of the battery performances.

The second area is about the propagation of the degradation parameters. In this phase, the run-to-failure history of a known battery is cooped with the new hidden parameters, identified at each cycle during the calibration phase, in the MLP-PF, whose details are described in Section 2.2. The hidden parameters are hence propagated in time and their Probability Density Functions can be computed (PDF) in each future instant of time.

Finally, in the last phase, the PDFs are used to determine the RUL of the battery, defined as the number of cycles in which the battery can provide the required power, and to estimate its future performances, simulating the model in a Monte Carlo approach.

2.1. Battery electrochemical model

The electrochemical behaviour of the battery has been derived following (Daigle & Kulkarni, 2015), with the goal of simulating the output voltage of a cylindrical cell with a nominal voltage of 4.2V. The model is built by use of Modelica (Modelica, 1996), a language well suited for designing 0D/1D models which, through the functional mock-up interface (FMI) standard (*Functional Mock-Up Interface*, 2010), allows the easy integration with other programming languages for the statistical analysis and the quantification of

uncertainties; This offers two advantages: first write directly the equations, in contrast with the typical variable assignment of a programming language, and secondly to use the model as a function, defining inputs and obtaining the corresponding outputs as results of the simulation.

The interested reader can refer to (Daigle & Kulkarni, 2013) and (Cancelliere & Girard, 2022) for a deeper description of the model and its equations. The main processes leading to the degradation of the cell over time are:

1. solid electrolyte interface (SEI) growth: formation of a solid layer on the surface of active material, which can happen both during cycling and storage at high temperatures.
2. lithium corrosion: lithium corrodes, leading to an irreversible loss of mobile ions.

In our model these processes are governed by the parameters q_{Max}, the number of lithium ions moving through the electrolytes during charges and discharges, and R_0, the internal resistance. The variation of these parameters, as well as the diffusion coefficient D, allows us to describe the performances of the cell during its lifetime.

However, these parameters are not directly measurable during usage. Hence, we need a method to infer them from the measurements available from the BMS. This work uses the NASA battery dataset (Saha & Goebel, 2007) to calibrate the model. In particular the battery denominated "B0005" is used as the training set. To obtain the parameters at the beginning of the battery's life, the output voltage of the first cycle is taken as a reference. To minimize the error between the actual curve and the model, we apply COBYLA, a numerical optimisation method for constrained problems. In this way, we obtain the optimal parameters that describe the behaviour of the battery at the beginning of its life.

This same algorithm, as can be appreciated in Figure 1 in the calibration area, is also used online to infer the degradation parameters during usage. Indeed, to account for the external changing condition and different usage modes, the parameters are updated online every time a new measurement is available. This enhances the reliability of the model and the accuracy of the predictions, which in turn improves the effectiveness of the maintenance strategy. Figure 2 shows the results of the calibration of the model at the beginning and at the end of the battery's life.

2.2. Multi layer perceptron particle filter

The electrochemical model developed, in combination with the methodology used to update online the degradation parameters, is a useful tool in monitoring the battery performances and identifying possible faults, hence aiding in the diagnostic. However, it still lacks information regarding the prediction of the RUL, which is defined as the remaining

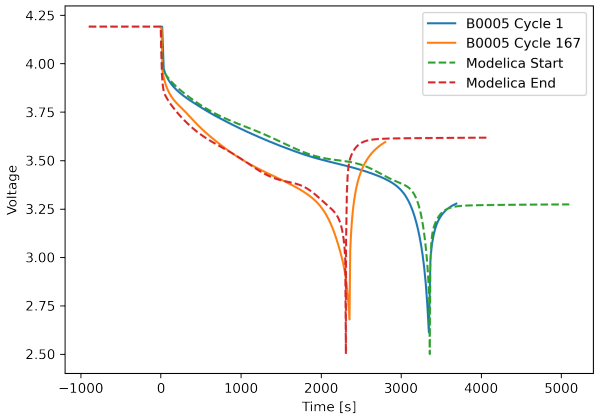

Figure 2. Calibration of the model degradation parameters at the beginning and end of battery's life. The variation of the degradation parameters produces a decrease in the time to discharge and an increase of the equilibrium potential, which the model is able to simulate.

number of cycles at which the battery can provide the requested power. To do so, it is necessary to evaluate the model with a prediction of the hidden degradation parameters, taking into account their variability due to the electrochemical nature of the problem. As reference, a run-to-failure history of the three parameters can be derived inferring them with the COBYLA algorithm on the entire life of the reference battery "B0005", as plotted in Figure 3.

However, as already pointed out, the specific battery can evolve very differently from the reference one. To account for that, the parameters are propagated with a multi-layer perceptron particle filter (Cadini et al., 2019), a methodology in which the weights and biases of an MLP are recurrently identified by a PF based on the online measurement coming from the BMS. A brief description of this methodology is now reported. For a thorough investigation of the functioning of an MLP and PF respectively, the reader can refer to (Doucet,

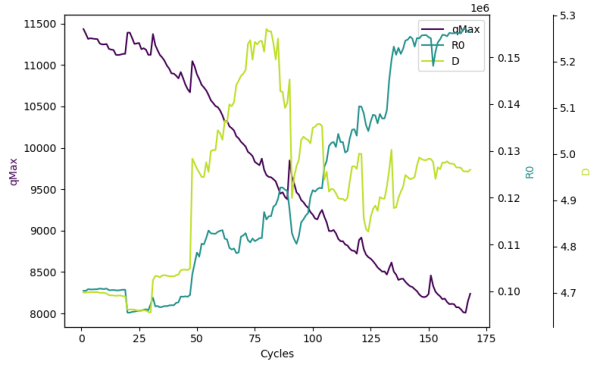

Figure 3. Run-to-failure behaviour of the three parameters for the battery "B0005". Each triple of values represents the optimal parameters found for each cycle.

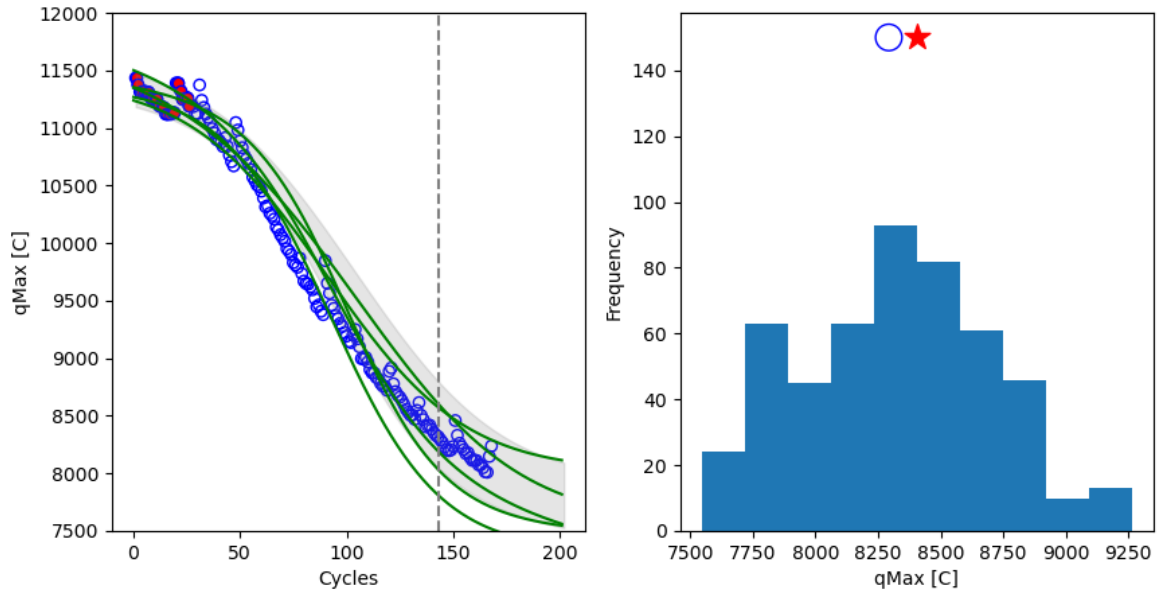

Figure 4. Left graph: propagation of the q_{Max} parameters through the MLP networks. Each green line represents a particle, the grey area contains the 95 percentiles of the overall particles. The red dot represent the available observation, the blue circles the future ones, not known to the algorithm. Right graph: PDF of the parameter at future cycle 143 (grey dotted line in the left graph). The blue circle represent the true value, the red star the mean of the PDF.

Godsill, & Andrieu, 2000) and (Arulampalam, Maskell, Gordon, & Clapp, 2002), while the particle filtering-based neural network training is detailed in (Sbarufatti, Corbetta, Giglio, & Cadini, 2018).

The three degradation parameters to be propagated are assumed to be independent, hence the methodology is applied to each of them separately. For brevity's sake, the methodology applied to q_{Max} is here shown, but the same reasoning holds for the other degradation parameters.

The structure of the MLP used in this work consists of a network with 1 input node, 1 hidden layer with 3 neurons and a single output node, thus with a total of 10 weights and biases. The use of a simple network is motivated by the necessity of having a relatively small number of weights and biases, which will have to be identified online by the PF. A bigger number of layers, and/or neurons per layer, from one point of view can improve the accuracy of the prediction, but on the other hand its cost is too high for the online purpose of this work.

The neural network is first trained on the historical data of q_{Max}, shown in Figure 3, which represent the expected trajectory. The network weights and biases are organized in a vector $x_0 = [x_1, x_2, ..., x_{10}]$, which represents the starting point of the process. Assuming the observation z_k, which in this case is the parameter q^k_{Max}, is available at the discrete time step k, we can describe the process and the measurement equations as:

$$x_k = x_{k-1} + \omega_{k-1} \tag{1}$$

$$z_k = g(x_k, k) + \eta_k \tag{2}$$

where $g(x_k, k)$ is the MLP network, taken as the model measurement equation, i.e. the relationship between the hidden states x_k and the observation z_k. ω_{k-1} and η_k represent the process and measurement noises respectively, both assumed Gaussian.

The task of the particle filter is to recursively estimate the probability density function $p(x_k|z_{0:k})$ which, from a Bayesian perspective, provides the degree of belief in the hidden degradation state x_k based on the previous knowledge, which are the measurements $z_{0:k}$. To estimate this PDF we recur to the sampling importance resampling (SIR) PF algorithm, in which N_s random state trajectory (particles) are generated on the basis of 1. To every particle is then assigned a weight based on its likelihood $p(z_k|x^{(i)}_k)$ every time a new observation z_k is available, representing the probability of observing z_k given the particle $x^{(i)}_k$. Finally the particles are re-sampled to avoid the sample impoverishment problem (Doucet et al., 2000): the particles with lower weights are more likely to be discarded, thus converging through the particles with higher weights.

This approach guarantees the adaptability to the real trend of the measurements and allows to compute the PDF in each

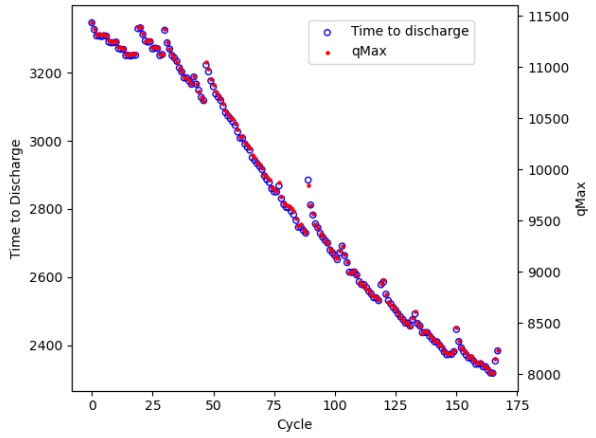

Figure 5. Correlation between time to discharge and the optimal parameter q_{Max} for battery B0005

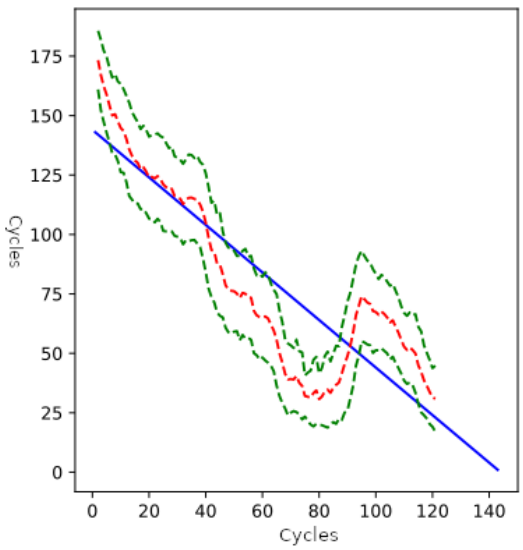

Figure 6. Remaining useful life prediction for the battery at cycle 125. The blue line represent the true battery RUL. The dotted lines represent the algorithm predictions, where the red line is the mean value and the green line are the 5^{th} and 95^{th} percentiles.

future instant of time, as shown in the left graph in Figure 4: the red dots represent the available observations, while the blue dot the future trend (not known to the algorithm). Each green line represents a particle, hence one of the $N_s = 500$ MLP network prediction of q_{Max}. With this approach it is possible to compute the posterior PDF of the parameter at each future cycle k, as illustrated in the right graph of Figure 4.

2.3. Remaining useful life estimation

In order to predict the RUL of the battery, we need to define a criterion for its end of life (EOL). A battery is usually said to reach the EOL when its capacity drops below 80% of the initial one, which is strictly correlated in our case (meaning constant discharge current) to the time to discharge (TTD). In order to exploit in the best possible way the available dataset, the authors decided to fix the threshold at $24000s$, hence with a remaining capacity of about 70%.

Moreover, as already stated in (Nascimento et al., 2021) and shown in Figure 5, there is an almost perfect correlation between the TTD and q_{Max}. This means that is possible to shift the threshold directly to this parameter, thus greatly simplifying the task of estimating the RUL, since there is no need to evaluate the physical model in order to obtain the TTD. Hence, for the purpose of our work, the EOL is defined as the cycle at which the parameter q_{Max} drops below the value of 8300 C.

Hence it is possible to compute the EOL_k^i for each MLP prediction i at the current cycle k, and consequently the RUL_k^i as:

$$RUL_k^i = EOD_k i - k \tag{3}$$

which are used to build the posterior probability density function of the RUL_k, $\hat{p}(RUL_k|z_{1:k})$. Figure 6 shows the results of a simulation in which is reported the expected value of the RUL from the first to the 120^{th} cycle with respect to the true battery RUL (solid blue line).

Once identified the $\hat{EOL}$, representing the mean value of the posterior PDF $\hat{p}(EOL_k|z_{1:k})$, it is possible to compute the PDFs for each degradation parameters at $k = \hat{EOL}$. These PDFs are used to sample the degradation parameters to feed the electrochemical model in a classic Monte-Carlo fashion, allowing the evaluation of the performances of the battery at its EOL. Figure 7 shows the results of a Monte-Carlo simulation: the red dotted line represents the true battery output voltage, while each grey line is a Monte-Carlo simulation. The blue line represents the average value, the green line its variance.

3. CONCLUSIONS

This paper proposes a combination of a physical model with a data-driven approach for the prognosis of lithium-ion batteries. The use of the physical model guarantees a better understanding of the underlying degradation dynamics, which is usually the main drawback of the pure black-boxes approaches. On the other hand, the integration with the data-driven algorithm allows for the propagation of the model parameters and the quantification of the uncertainties involved. The use of Modelica, with its declarative approach in defining the physical model, provides a more intuitive description of the physics and simplifies the modelling process. Moreover, the integration through the functional mock-up inter-

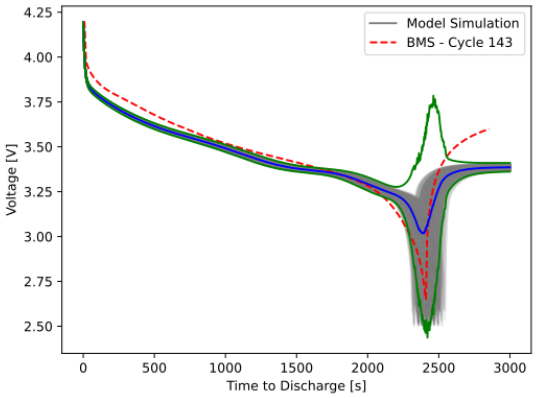

Figure 7. Monte-Carlo simulation of the battery performances at the EOL with the parameters estimated at current cycle $k = 125$ for future cycle $k = 143$, identified as battery EOL.

face in Python provides a powerful and flexible platform for parameter optimisation, sensitivity analysis, and other analysis tasks, as evidenced in this work with the optimisation of the degradation parameters. The results of the simulations show good agreement with the actual battery performance, indicating that the proposed approach could be a useful tool for predicting RUL and evaluating battery health in real-world applications.

Future work on this matter will include the use of different discharging profiles, closer to a real-life application, and the relaxation of the hypothesis on the independence between the degradation parameters.

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