Predicting Lithium-Ion Battery Degradation Modes Using a Machine Learning Approach Based on EIS Measurements

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***Abstract*—This study presents a machine learning (ML) framework that is able to predict the degradation modes of a lithium-ion battery. Leveraging on some analytical models recently proposed in literature, the proposed algorithm estimates three main ageing indicators (Conductivity Loss (CL), Loss of Active Material (LAM) and Loss of Lithium Inventory (LLI)), using ML models based on battery-related features, i.e., Electrochemical Impedance Spectroscopy (EIS), State of Charge (SoC), Open Circuit Voltage (OCV) and number of cycles. In order to test the accuracy of these ML models, two different datasets were created (for testing and validation), using data acquired from 8 different cells. Experimental results show that the proposed approach, based on ML, is able to determine the ageing status of a battery with high accuracy, thus offering a reliable solution for battery health diagnostics.**

***Index Terms*—Battery Management System (BMS), Machine Learning, Random Forest, Battery State of Health (SoH)**

Nowadays, lithium batteries are a key technology for numerous applications, from consumer electronics to electric vehicles and energy storage systems. However, the progressive degradation of their performance over time limits their reliability and operating life, making fundamental the development of models able to accurately estimate their state of health (SoH) and remaining useful life (RUL).

Unfortunately, the development of analytical models that are able to estimate the ageing status of a battery is a very difficult task, as it depends on several internal or external effects (such as changes over time of porosity and conductivity, chemical reactions, changes of the structure, electrode degradation) that are quite difficult to be analyzed and modeled [1] [2].

In [3] the authors identified three main ageing indicators that could be estimated to determine the state of a battery:

- the **Conductivity Loss** (CL), which includes the degradation of the electronic parts of the battery such as the corrosion of current collectors or the degradation of binder;
- the **Loss of Active Material** (LAM), which includes the structural changes in the active material and the decomposition of the electrolyte;
- the **Loss of Lithium Inventory** (LLI), which includes the changes in the number of available lithium-ions (Li-ions).

In [3] [4], authors proposed analytical models able to estimate CL, LAM and LLI. These algorithms firstly attempt to determine the parameters of an equivalent circuit model of the battery, starting from measures obtained using Electrochemical Impedance Spectroscopy (EIS) [5] or Incremental Capacity and Differential Voltage (IC-DV) [6] techniques. Later, the algorithms determine the values of CL, LAM and LLI as an analytic function of the parameters of the equivalent circuit of the battery. Normally, these models are subjected to a number of limitations. For example, IC and DV require slow signal acquisition, making them unsuitable for practical applications. Conversely, EIS circuit-fitting algorithms are complex, requires iterative extraction and are subject to inaccuracies and artefacts caused by human error. Furthermore, Li-ion batter-

TABLE I
TESTED CHARGING/DISCHARGING CYCLES

Cell	Ageing profile	Discharge	Galvanostatic Charge	Potentiostatic Charge	Temp.
1	A	5C (15A)	1.4C (4A)	up to 0.15C	25 °C
2	A	5C (15A)	1.4C (4A)	up to 0.15C	25 °C
3	B	5C (15A)	1.7C (5A)	up to 0.15C	25 °C
4	B	5C (15A)	1.7C (5A)	up to 0.15C	25 °C
5	C	5C (15A)	1.4C (4A)	up to 0.15C	45 °C
6	C	5C (15A)	1.4C (4A)	up to 0.15C	45 °C
7	D	5C (15A)	1.4C (4A)	none	25 °C
8	D	5C (15A)	1.4C (4A)	none	25 °C

ies are subjected to several internal degradation phenomena (electrode degradation, electrolyte decomposition, oxidation), which complicate the right extraction of equivalent circuit model parameters and the accuracy of the outcomes.

An alternative solution may be the usage of machine learning (ML) models. Several authors [7] [8] [2] demonstrated that ML is a promising solution for state of health (SoH) battery estimation. These algorithms generate a model of the battery using a training dataset that consists of several input-output pairs of measures previously acquired on the device [9]. Conventionally, these algorithms do not use an equivalent circuit of the battery and do not require a knowledge of the internal electrochemical processes of the device.

Differently from previous works, this one investigates the performance of ML algorithms specifically devised to determine the ageing status of a lithium-ion (Li-Ion) battery. The algorithms were trained, tested and validated using a dataset provided by CNR ITAE.

More in details, CNR ITAE provided the EIS-Data-Ageing (EISDA) dataset, which contains data collected from 8 cylindrical lithium-ion cells (model Samsung INR18650 30Q, 2.95 Ah). These batteries, hereinafter identified with integer numbers from 1 to 8, were aged under four different cycling profiles (varying temperature, charge current, and depth of charge, see Tab. I for further details).

The main contribute of this paper is an assessment of the accuracy of two ML algorithms (Random Forest, RF [10] and Support Vector Regression, SVR [11]) in the ageing estimation of CL, LAM and LLI when they are applied on EISDA dataset.

The paper is organized as follows. Section I deals with the related works. Section II deals with the model used to describe the degradation phenomena occurred in the battery. Section III describes the EIS dataset used to train the proposed ML algorithms and reports the experimental results. Finally, Section IV deals with conclusions.

I. Related works

As observed in [8], the algorithms for the estimation of ageing state of a battery can be divided in two classes: model-based and model-less.

Many model-based algorithms exploit an electrochemical model (ECM) of the battery. Parameters of the model are determined from experimental data using fitting methods. For instance, in [3] [4] the authors proposed a technique to estimate CL, LAM and LLI using an ECM model.

The model-less methods regard the system as a black box. An algorithm is used to generate a model directly from the features extracted from the battery. ML-based, statistical and computational algorithms are examples of model-less methods.

Several authors reported promising results in SoH estimation, using ML algorithms. As for example, SoH models were generated using Neural Networks (NN) [12], Support Vector Regression (SVR) [13] or Random Forest (RF) [2], [14].

Conversely, in literature there are few papers that deal with algorithms able to estimate the level of CL, LAM and LLI degradation using a model-less approach.

As for example, in [15] Lee used an artificial neural network (ANN) to estimate LAM and LLI. In [16] Chen used an algorithm based on RF and SVR to estimate the level of LAM degradation, but the method required a pre-classification of the batteries in order to determine the main reason of ageing (LLI, LAM or solid electrolyte interphase (SEI) formation).

At the best of authors knowledge, this is the first paper where the performance of RF and SVR in generating models for ageing estimation are investigated considering all the three indicators (CL, LAM and LLI) at the same time.

It is worth noting that methods based on RF and SVR are particularly suitable for battery management systems (BMSs) as they require less computational effort than other solutions based on neural networks [1].

II. Battery model

The identification and quantification of degradation phenomena in lithium-ion cells was carried out by adapting an analytical model previously reported in the literature [3] [4]. This model represents the three main ageing indicators of a battery (CL, LAM and LLI), as a tuple of percentage values ($CL_q\%$, $LAM_q\%$ and $LLI_q\%$ respectively, where q is the specific SoC level of the cell).

In the proposed model, each degradation phenomenon was correlated with parameters from an equivalent circuit model (shown in Fig. 2), specifically designed to represent the typical impedance spectrum of a lithium-ion cell.

It is worth noting that although the equivalent circuit used for fitting EIS data consists of seven circuit elements (L, R_{ohm}, CPE_1, R_{ct1}, CPE_2, R_{ct2}, Z_w), it is mathematically described by nine parameters, since the impedance of each Constant Phase Element (CPE) is expressed using two terms: Q and α (being $Z = 1/Q * (j\omega)^{\alpha}$).

However, the model used for degradation identification utilizes only four of these nine parameters, specifically those representing the real part of the impedance: R_{ohm}, R_{ct1}, R_{ct2}, and Z_w. The remaining elements (L, CPE_1, and CPE_2) are still essential for achieving an optimal fit of the EIS spectra, which is necessary for accurately extracting the four parameters of interest.

The absolute quantification of the three degradation phenomena (CL, LAM, and LLI) is performed by calculating the percentage variation of each of the four relevant circuit

Fig. 1. Arbin multichannel cycler

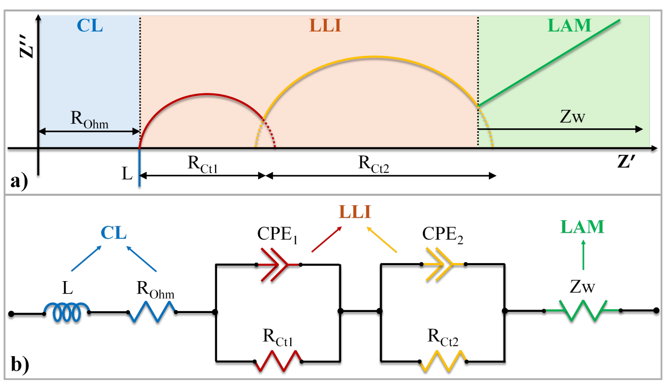

Fig. 2. Equivalent Circuit for EIS Spectrum fitting

parameters (or a combination of R_{ct1} and R_{ct2} in the case of LLI) over the aging cycles (k), relative to the initial total cell resistance ($R_{tot(0,q)}$ at k = 0).

The three degradation metrics, $CL_q\%$, $LAM_q\%$, and $LLI_q\%$ are mathematically defined by Eqs. 1-3. The initial total resistance ($R_{tot(0,q)}$) is defined as the sum of the four aforementioned parameters, as shown in Eq. 4, where q represents the SoC at which the cell resistance is measured. It is important to note that $R_{tot(0,q)}$, and more generally the impedance of a lithium-ion cell, depends on the specific SoC at which it is measured. In other words, for a fixed number of aging cycles, the parameters of the equivalent circuit used for fitting vary, as they depend on the SoC.

$$CL_q\% = 100 \cdot \frac{R_{ohm(0,q)} - R_{ohm(k,q)}}{R_{tot(0,q)}} \tag{1}$$

$$LAM_q\% = 100 \cdot \frac{Z_{w(0,q)} - Z_{w(k,q)}}{R_{tot(0,q)}} \tag{2}$$

$$LLI_q\% = 100 \cdot \frac{(R_{ct1(0,q)} - R_{ct1(k,q)}) + (R_{ct2(0,q)} - R_{ct2(k,q)})}{R_{tot(0,q)}} \tag{3}$$

$$R_{tot(0,q)} = R_{ohm(0,q)} + R_{ct1(0,q)} + R_{ct2(0,q)} + Z_{w(0,q)} \tag{4}$$

As a result, the overall quantification of cell aging and thus of the three associated degradation phenomena ($CL_q\%$, $LAM_q\%$, and $LLI_q\%$) depends on the SoC and ageing cycle at which the measurement is taken. Since the impedance of the cell at different SoC levels is more or less correlated with physical-chemical phenomena occurring at the anode or cathode, this type of analysis at multiple SoC levels may allow for further discrimination of degradation mechanisms, potentially associating them separately with the anode or cathode. Finally, since LAM and LLI are primarily related to the availability of active lithium ions within the cell, their relative percentages can be used to quantify their respective contributions to capacity loss. Similarly, since CL is associated with the ability of the cell to deliver current, an increase in its value reflects a loss in power release. Through this ageing characterization, the EIS spectra were correlated with the three main ageing mechanism and, so each spectrum is "tagged" by them. The extraction of the parameters has been done through an automated and innovative fitting procedure based on a Python code that is not the focus of this work. However, the outcomes of this extraction procedure are considered the input of the machine learning algorithm proposed. It is worth noting that the "tagging" process could be differently performed and the ML model developed is not dependent on it.

The overall approach adopted in the current work is summarized in the concept of Fig. 3. The main difference with traditional models based on EIS parameters extraction is the use of the existing correlation between circuit parameters and the three ageing phenomena as input for the machine learning. More specifically, after the initial extraction of circuit parameters from EIS experimental data and the training of the ML model according to the input variables selected, the proposed ML algorithm outputs an estimation of LAM, LLI and CL, thus eliminating the need to further fit the EIS data to a circuit model. As a primary consequence, it results in a significant reduction in modeling time. This was achieved through a fully automated process that does not require additional manual intervention or iterative optimization but only the right identification of the EIS spectrum coming from the in-situ measurement and associated ageing phenomena.

Moreover, it is worth noting that the proposed approach differs from other algorithms proposed in literature [17], where a neural network is used to determine the parameters of the

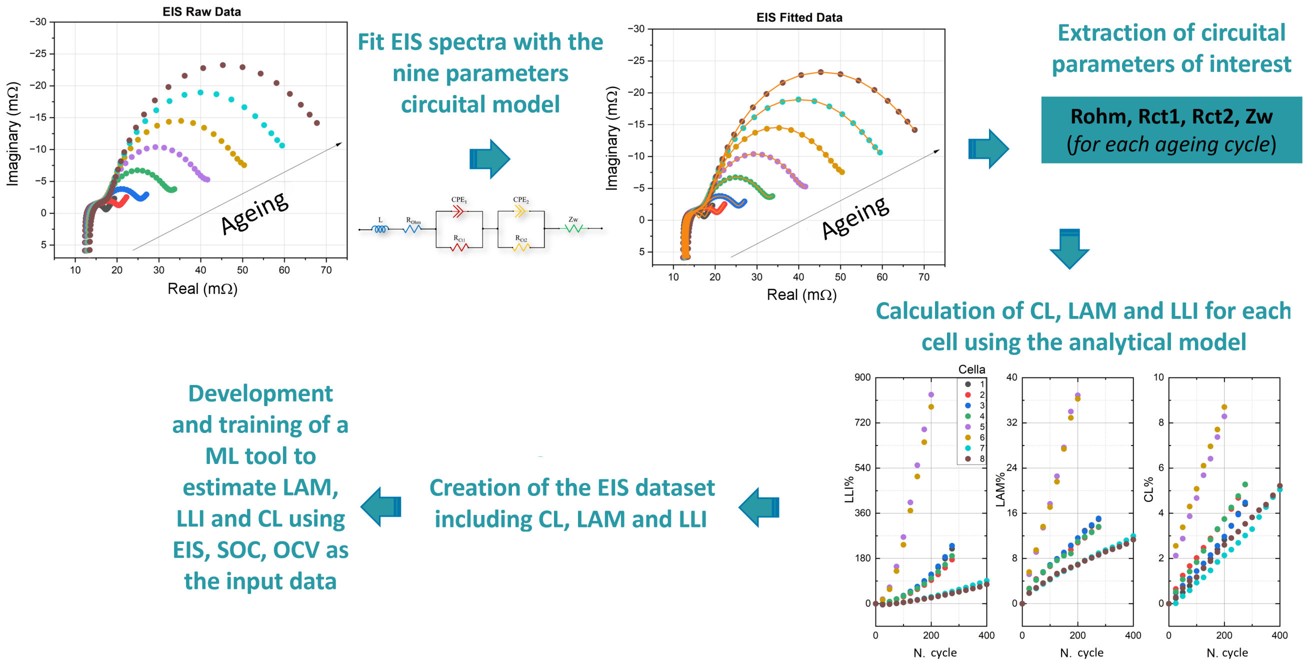

Fig. 3. Procedure used to generate the model

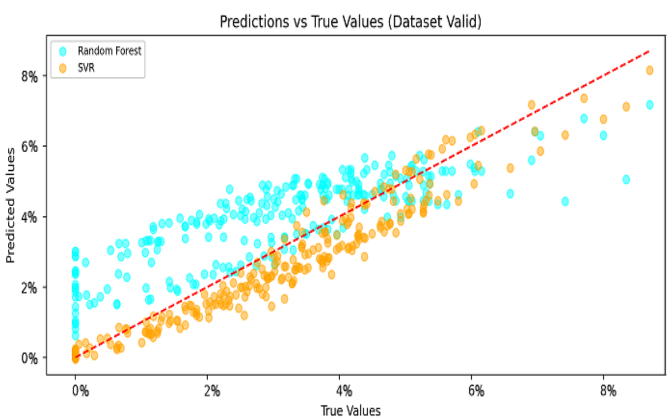

Fig. 4. Concordance plot comparing the performance of RF and SVR models in $CL_q\%$ estimation (5 features).

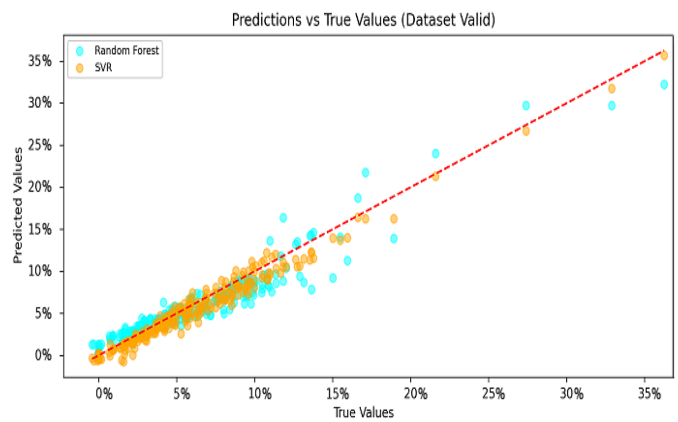

Fig. 5. Concordance plot comparing the performance of RF and SVR models in $LAM_q\%$ estimation (5 features).

equivalent circuit of the battery. In these papers the values of LAM, LLI and CL could be estimated only applying in a second step the Eqs. 1-3 to the output of the ML algorithm.

III. Experimental results

All the experimental results were obtained using the EIS-Data-Ageing (EISDA) dataset provided by CNR-ITAE.

The dataset represents different features of the 8 batteries (Samsung INR18650 30Q) under test. In particular, each row of the dataset contains a set of 105 features: the Ageing cycle, the State of Charge (SOC), the Open Circuit Voltage (OCV) and 51 EIS impedance values (totally 102 features considering module and phase). The features were acquired during the lifetime of the battery at different ageing cycles until the battery reached 80% of the initial capacity.

More specifically, EIS measurements were taken every 25 cycles at five SoC levels (100%, 75%, 50%, 25%, 0%), in the frequency range of 0.1 Hz to 10 kHz, with 10 points per decade, and an excitation source of 0.01 V. All measurements were done with the use of an Arbin LBT21044 multichannel cycler, which provides different mini-climatic chambers for each cell and direct integration of a Gamry 5000E potentiostat for EIS spectra extraction.

The dataset was partitioned into two subsets. The first one, named EISDA-1357, contained data related to the odd cells, while the second one, named EISDA-2468, contained data related to the even cells.

EISDA-1357 was used to perform a 5-fold cross-validation [18]. EIS-1357 dataset has been partitioned into 5 folds (i.e., 5 equal-sized parts) whose items were randomly selected. The model was trained and tested 5 times, each time using a differ-

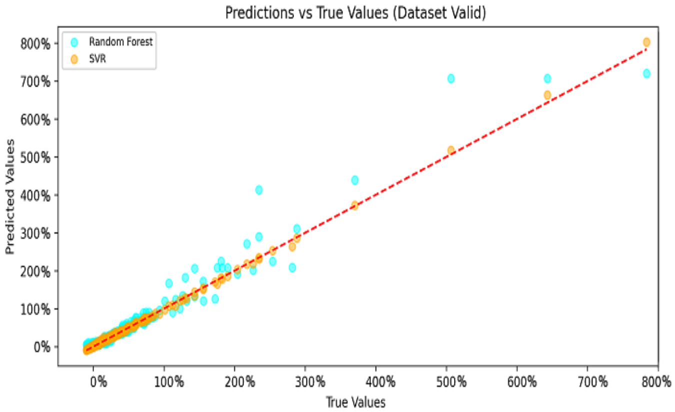

Fig. 6. Concordance plot comparing the performance of RF and SVR models in $LLI_q\%$ estimation (5 features).

TABLE II
CROSS-VALIDATION RESULTS (RF REGRESSOR)

	$CL_q\%$		$LAM_q\%$		$LLI_q\%$	
Model	RandomForest (e=10;d=5)		RandomForest (e=10;d=5)		RandomForest (e=10;d=5)	
Dataset	EISDA 1357 TR20	EISDA 2468	EISDA 1357 TR20	EISDA 2468	EISDA 1357 TR20	EISDA 2468
MSE	0.07%	1.83%	1.16%	2.21%	61.24%	512.52%
MAE	0.23%	1.11%	0.9%	1.06%	5.91%	10.34%
MXABE	0.62%	3.28%	2.25%	5.8%	24.18%	200.16%

ent fold as the test set (EISDA-1357-TR20) and the remaining 4 folds as the training set (EISDA-1357-TR80). Finally, the whole dataset EISDA-2468 was used for validation.

The performance of each ML model was measured, over the subset used for testing or validation, by calculating the mean squared absolute error (MSE), the mean absolute error (MAE) and the maximum absolute error (MXABE), defined as follows:

$$MSE = \frac{\sum_{i=1}^{n}(y_i - \hat{y}_i)^2}{N} \tag{5}$$

$$MAE = \frac{\sum_{i=1}^{n}|y_i - \hat{y}_i|}{N} \tag{6}$$

$$MXABE = max_{i=1..n}|y_i - \hat{y}_i| \tag{7}$$

where y_i are the actual values of the ageing indicator under analysis, and $\hat{y}_i$ are the estimated values of the same indicator obtained by the ML model.

A first experiment was carried out, aimed to determine the performance of RF models in estimating the $CL_q\%$, $LAM_q\%$ and $LLI_q\%$ values.

As the performance of RF-based models depends on the number of decision trees (e) and on their maximum depth (d) [2], a preliminary analysis was performed in order to determine the optimal values of these parameters. The analysis showed that the RF models achieved the best performance using $e = 10$ decision trees with $d = 5$ maximum depth.

The average MSE, MAE and MXABE values, calculated over the 5 partitions with the optimal parameters, are shown in Tab. II. The experimental results show the MAE in the estimation of $CL_q\%$ and $LAM_q\%$ on the testing dataset was about 1%, whereas the MAE in the estimation of $LLI_q\%$ was one order of magnitude higher. Moreover, also the MAXABE in estimating $LLI_q\%$ was very high (200.16%), thus leading to the conclusion that RF models are unsuitable for a reliable estimation of battery ageing.

TABLE III
CROSS-VALIDATION RESULTS (SVR REGRESSOR)

	$CL_q\%$		$LAM_q\%$		$LLI_q\%$	
Model	SVR (C=2000; eps=0.1; kernel='rbf')		SVR (C=10000; eps=0.01; kernel='rbf')		SVR (C=100000; eps=1; kernel='rbf')	
Dataset	EISDA 1357 TR20	EISDA 2468	EISDA 1357 TR20	EISDA 2468	EISDA 1357 TR20	EISDA 2468
MSE	0.01%	0.45%	0.13%	0.97%	1.28%	11.71%
MAE	0.1%	0.56%	0.23%	0.78%	0.9%	2.22%
MXABE	0.31%	1.53%	1.27%	2.7%	3.39%	19.69%

A second experiment was performed, aimed to determine the performance of SVR models in estimating the $CL_q\%$, $LAM_q\%$ and $LLI_q\%$ values.

Also in this case, a preliminary test was performed in order to determine the best set of parameters (kernel type, regularization parameter C, and $epsilon$ (eps) i.e., the width of the no-penalty tube used by the SVR training loss function) that should be passed to the SVR regressor [19] [20] in order to achieve the best performance. The test showed that the SVR regressors for the estimation of $CL_q\%$, $LAM_q\%$ and $LLI_q\%$ values require to be configured using different parameter sets (C and $epsilon$) to achieve the optimal results. The MSE, MAE and MXABE values achieved by the SVR regressors and the optimal parameters used for configuration are shown in Tab. III.

A comparison between Tab. III and Tab. II shows that the SVR regressors provided better results than RF regressors. In particular, the errors related to $LLI_q\%$ were reduced of one order of magnitude.

Fig. 4, 5 and 6 show the concordance plots related to $CL_q\%$, $LAM_q\%$ and $LLI_q\%$ respectively, thus allowing to compare the accuracy of RF and SVR models.

IV. CONCLUSIONS

In this paper we proposed three ML models for estimating three well-known indicators of the ageing state of a lithium-ion battery: Conductivity Loss (CL), Loss of Active Material (LAM) and Loss of Lithium Inventory (LLI).

The ML models, based on RandomForest (RF) and Support Vector Regression (SVR) algorithms, were trained using a dataset containing 105 features related to 8 batteries (State of Charge (SOC), Open Circuit Voltage (OCV) and EIS impedance values), acquired at different ageing cycle.

Once trained, the models are effective in providing an estimation of LAM, LLI and CL values of an unknown ageing status of the battery, starting from an input vector containing the EIS features of the storage system.

Differently from other approaches already presented in literature, the proposed method does not require a pre-classification of the batteries in order to determine the main reason of ageing. Moreover, the trained models work without fitting the input EIS data to a circuit model.

The performance metrics of the three RF and SVR regressors for LAM, LLI and CL estimation were measured using a 5-fold cross-validation procedure.

The experimental results show that SVR regressors provided better results than RF regressors. In particular, the MAE values achieved by the SVR regressors were equal or lower than 2.22%, whereas the MAE values achieved by the SVR regressors were equal or lower than 10.34%.

Future research activity will be focused on the implementation of the method presented in this work in the firmware of a battery management system, in order to test and validate the performance in a dynamic scenario.

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