

A fast fuel cell parametric identification approach based on machine learning inverse models

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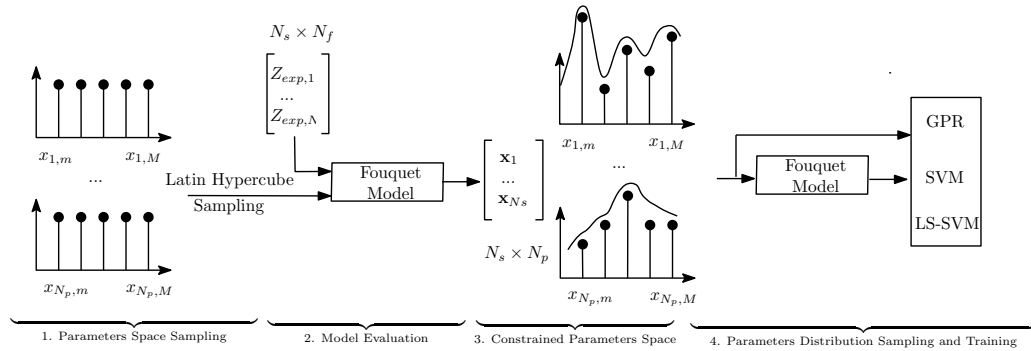
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(Article begins on next page)

1 Graphical Abstract

2 **A fast Fuel Cell Parametric Identification approach based on Machine Learning Inverse Models**

4 Antonio Guarino, Riccardo Trincherò, Flavio Canavero, Giovanni Spagnuolo



5 Highlights

6 **A fast Fuel Cell Parametric Identification approach based on Ma-**
7 **chine Learning Inverse Models**

8 Antonio Guarino, Riccardo Trincherò, Flavio Canavero, Giovanni Spagnuolo

- 9 • Parameter identification of equivalent circuit model performed through
10 Inverse Model;
- 11 • Parameter search space is first constrained in feasibility regions;
- 12 • Inverse Models built through GPR, SVM and LS-SVM machine learn-
13 ing regression models;
- 14 • The identification results are shown for both synthetic and experimen-
15 tal data;
- 16 • Performance and Resource Usage of the embedded implementation are
17 explored.

18 A fast Fuel Cell Parametric Identification approach
19 based on Machine Learning Inverse Models

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22 **Abstract**

In this paper a computationally efficient optimization approach to the parametric identification of a fuel cell equivalent circuit model is presented. It is based on the inverse model and on machine learning regressions. During the training phase, the inverse model is built numerically by means of advanced regression approaches, i.e., the support vector machine regression, the least squares support vector machine regression and the Gaussian process regression. The training set is synthetically generated to the aim of exploring the parameter space and to characterize different stack operating conditions, including normal and faulty ones. The accuracy of the considered approaches is investigated by employing a test set including many experimental data, consisting of impedance spectra measured through the electrochemical impedance spectroscopy and referring to very different stack operating conditions. The results show that all the considered machine learning methods allow to identify the parameters of the fuel cell model with a low computational burden, so that they fit with the hardware resources of low cost embedded processors. This feature allows to envisage that the proposed approaches are good candidates for a model-based on-line diagnosis of fuel cell stacks.

23 *Keywords:* Parametric identification of Equivalent Circuit Model, Inverse
24 Model, Machine Learning Regression

25 **1. Introduction**

26 Fuel Cells (FCs) and hydrogen-related technologies are a key point of the
27 European Green Deal and of the Recovery Plan. In order to improve the

28 Life Cycle Assessment (LCA) of FCs' stacks, a significant lifetime has to be
29 reached. Monitoring and Diagnostic Tools (MDTs) are helpful in evidencing
30 a malfunctioning and the occurrence of operating conditions that stress the
31 stack and reduce its Remaining Useful Life (RUL). Although many diagnos-
32 tic approaches and methodologies are proposed in literature, very often they
33 employ laboratory measurement systems and/or algorithms requiring signifi-
34 cant hardware resources. Consequently, their on-line and on-site application
35 becomes very challenging. At the same time, some efforts are on course to
36 develop MDTs employing hardware systems and requiring computational ef-
37 forts that are compatible with FC systems costs, diagnostic accuracy and
38 time response. For instance, some H2020 funded projects, e.g. [1], have been
39 aiming at pursuing the objective of having an Electrochemical Impedance
40 Spectroscopy (EIS) -based MDT using ordinary power electronics and em-
41 bedded processors to perform frequency domain measurements and detect
42 a number of faults through on-line diagnostic algorithms. The diagnostic
43 methods usually work in two spaces: one is the frequency domain, i.e., the
44 stack impedance spectrum in the Bode or in the Nyquist planes, and the
45 other is the multidimensional space of the parameters of a suitable non lin-
46 ear FC stack Equivalent Circuit Model (ECM). The latter requires a robust
47 approach that takes the impedance spectrum as an input and gives at the
48 output the set of ECM parameters. This task is quite challenging because of
49 the number of parameters, of the wide search space and of the lack of a good
50 guess solution. A further issue is the high non linearity between the function,
51 which is usually considered as the objective of the optimization problem, e.g.,
52 the Root Mean Square Error (RMSE) between the ECM impedance and the
53 experimental data, and the parameters thereof. Some approaches are pro-
54 posed in literature, e.g. in [2], but they are based on specific assumptions
55 about the signal-to-noise level affecting the data; consequently, they do not
56 always ensure the convergence towards the absolute minimum RMSE value
57 or they have a significant computational burden.

58 Machine learning (ML) has been using more and more in energy problems.
59 An increasing number of ML-based approaches to monitoring, diagnosis and
60 RUL prediction of batteries and FCs can be found in the recent literature. In
61 [3, 4] multiple ML-based approaches have been employed to RUL estimation
62 of a set of batteries by using charging, discharging, internal resistance and
63 temperature data. The effectiveness of such approaches that operate on
64 the battery early usage data is demonstrated. In [5], the battery state of
65 health is estimated by employing mechanical and electrical features through

66 an Autoencoder and a Gaussian Process Regression (GPR) based estimation
67 model. In [6, 7] an artificial neural network is used to estimate the remaining
68 battery capacity. In [8], a number of variables, e.g., stack current and voltage,
69 air supplier control signal, temperatures, air flow rates, is monitored and their
70 values are compared with those ones computed through a suitable model.
71 The patterns of the resulting differences are used to train supervised ML
72 methods that are then able to classify different faults. In order to ensure
73 a simultaneous faults detection, instead then addressing independent faults
74 only, in [9] a deep learning network called stacked sparse autoencoder is
75 proposed. The stack voltage is also the subject of the study presented in [10]:
76 some relevant features are extracted and allow ML approaches to identify the
77 fault.

78 FC degradation prediction in presence of a variable load is performed in
79 [11] and [12] through a method based on a Genetic Algorithm (GA) and an
80 Extreme Learning Machine (ELM). The long term dynamic behavior of the
81 stack voltage is exploited in [13] to predict the performance degradation by
82 using a Grid Long Short-Term Memory (G-LSTM) Recurrent Neural Net-
83 work (RNN). A Support Vector Machine (SVM) classifier is adopted in [14]
84 for detecting FC faults by using a 3D model. The validation of the multi-
85 scale hybrid degradation index formulated in [15] and of the the approach
86 aimed at predicting the FC RUL presented in [16] is done through a ML tool
87 operating on experimental data. Moreover, a ML algorithm has been used
88 in [17] to find the best trade off between the EIS measurement time and the
89 fault detection accuracy through the impedance spectrum. The diagnosis is
90 conducted in the frequency space only, without any parametric identification
91 process. To this regard, the adoption of ML techniques for system param-
92 eters identification is quite rare in literature. Few examples refer to dynamical
93 systems identification: e.g., in [18] the systems considered are described by
94 differential-algebraic equations and the sum of the squared deviations of the
95 values of the state vector's coordinates from their exact counterparts obtained
96 through measurements at different time instants is considered as the objec-
97 tive function whose value is minimized through the adaptive random search
98 method. In [19], transfer learning methods are used to address a challenge in
99 state-space linear parameter-varying model identification and learning. Ker-
100 nelized ML is adopted when the distributions of the training and testing sets
101 are different.

102 This paper shows that the parametric identification of a Polymer Elec-
103 trolyte Membrane (PEM) FC ECM can be effectively performed by using

104 the *inverse model* [20]. A mathematical model, i.e., a multivariate func-
105 tion, is achieved as first. Then, it exploits a set of experimental EIS spectra
106 to identify the parameters of the widely recognised ECM presented in [21].
107 Since the parameters identification is carried out via a straightforward eval-
108 uation of the inverse model over the experimental values of the EIS spectra,
109 the proposed approach ensures a good compromise between computational
110 burden and accuracy, thus a performance that is better than the one the
111 stochastic identification techniques presented in literature are able to guar-
112 antee [22][23][24].

113 The construction of the inverse model, which is the core of the proposed
114 method, is not straightforward. Indeed, the inverse map is usually extremely
115 non-linear and ill-posed, this meaning that a given EIS spectrum might be
116 generated by more than one set of the ECM parameters. In order to mitigate
117 the above issue, in this paper three advanced ML regression methods are
118 considered. Specifically, the SVM regression [25], the Least Squares SVM
119 regression (LS-SVM) [26] and the GPR [27] are adopted to construct the
120 inverse model from synthetic data. The above techniques have been selected
121 for two main reasons. First of all, they are non-parametric techniques in
122 which the number of unknowns to be estimated during the model training
123 is independent from the dimensionality of the input space, i.e., from the
124 number of frequency points in the EIS spectrum. This allows to minimize
125 the number of training samples required by the model training and thus the
126 model complexity. Moreover, the regularizer used by the SVM and LS-SVM
127 regressions, as well as the noise term in the GPR, can be used to mitigate the
128 ill-posedness of the inverse model [28, 29]. The proposed approaches have
129 been also implemented in a low cost embedded system to demonstrate their
130 on-site and on-line potential; their performance is validated through a large
131 test set consisting of experimental EIS spectra.

132 The experimental test set has been obtained during the HEALTHCODE
133 H2020 project [1] on a PEM stack having 46 cells and an active area of 100
134 cm^2 . The stack was tested at $0.4 A/cm^2$ under different hydrogen and oxygen
135 utilization and for different humidity levels. The considered dataset refers
136 to cells number 1, 2, 23, 45, and 46 with measurements performed in the
137 range from 50 mHz to 2 kHz for a total of 157 spectra. Further details on
138 the testing procedure and on the study of the impact of water management
139 and reactant utilization through EIS are disclosed in [30][31].

140 The paper is organized as follows. Section II is aimed at describing the
141 proposed ML-based identification approach and Section III gives the details

142 of the application of the ML methods adopted. Section IV shows the proce-
 143 dure used to produce the synthetic data that have been used for the learning
 144 phase. Sections V and VI provide the analysis of the results based on syn-
 145 thetic and experimental data, respectively. Section VII shows the results of
 146 an approach validation performed through an embedded platform that is a
 147 candidate for the on-line application of the proposed approaches. The com-
 148 putational burden of the proposed approach in comparison with that one
 149 of an accurate stochastic approach proposed in literature is also provided.
 150 Conclusions end the paper.

151 2. Gray-Box System Identification via Forward Models

System identification aims at building a mathematical model on the basis
 of measured data [32]. Given the experimental FC impedance values $\mathbf{Z}_{exp} =$
 $[Z_{exp}(f_1), \dots, Z_{exp}(f_{N_f})]^T \in \mathbb{C}^{N_f}$ in a given frequency range, the goal is to
 identify the optimal configuration \mathbf{x}_* of the $N_p = 6$ free parameters $\mathbf{x} =$
 $[x_1, \dots, x_{N_p}]^T$ of the Fouquet ECM $\bar{Z}_F(f; \mathbf{x})$ (see the Appendix and [21] for
 additional details), such that:

$$Z_{exp}(f) \approx \bar{Z}_F(f; \mathbf{x}_*), \quad (1)$$

152 Literature results allow to assume that the Fouquet model is able to fit, with
 153 a good accuracy, the experimental spectra.

154 The chosen ECM has a closed-form and it provides a gray-box model, i.e.,
 155 a model built from a-priori knowledge, for the system identification. The
 156 Fouquet model is a forward map, or forward model, that for any configuration
 157 \mathbf{x} of its parameters is able to provide the corresponding impedance value
 158 $\bar{Z}_F(f; \mathbf{x})$ at the desired frequency value f .

159 The identification problem (1) can be also formulated by means of an
 160 objective function whose value has to be optimized through a suitable al-
 161 gorithm. For instance, the configuration \mathbf{x}_* can be estimated as the one
 162 that minimizes the average of the magnitudes of the squared error computed
 163 between the experimental impedance values collected in \mathbf{Z}_{exp} and the cor-
 164 responding ones predicted by the Fouquet model \bar{Z}_F in a given frequency
 165 range:

$$\mathbf{x}_* = \arg \min_{\mathbf{x}} \frac{1}{N_f} \sum_{i=1}^{N_f} |Z_{exp}(f_i) - \bar{Z}_F(f_i; \mathbf{x})|^2. \quad (2)$$

166 The properties of such an optimization problem, e.g. its convexity, depend
 167 on the structure of the forward model \bar{Z}_F . As an example, the problem at
 168 hand turns out to be convex when the model \bar{Z}_F is so simple that it can be
 169 written as a linear combination of its input parameters \mathbf{x} . However, such
 170 minimization problem can be usually classified as a non-convex optimization
 171 with several local minima [33], which might require advanced optimization
 172 algorithms, as in [22][23].

173 Alternatively, provided that the candidate model $\bar{Z}_F(f; \mathbf{x})$ can be inex-
 174 pensively evaluated for any set of the input parameters \mathbf{x} , the above optimiza-
 175 tion problem can be tackled via a grid search approach. It is a brute force op-
 176 timization scheme, in which the N_p parameters can be interpreted as uniform
 177 distributed random variables in a given range (i.e., $x_i \sim \mathcal{U}(x_{i,min}, x_{i,max})$),
 178 then the model $\bar{Z}_F(f; \mathbf{x})$ is evaluated for a large number of realizations of the
 179 input random parameters drawn according to their probability distribution
 180 functions (PDFs). Thus, the optimal parameters set \mathbf{x}_* can be estimated
 181 among all the considered realizations as the one that minimizes the cost
 182 function of the optimization problem (2). This approach is straightforward
 183 and does not require to calculate the gradient function of the cost function,
 184 but it turns out to be quite cumbersome and computational expensive. In
 185 fact, the accuracy and the reliability of optimal solution \mathbf{x}_* predicted by such
 186 brute force approach heavily depends on the number of calls to the model
 187 \bar{Z}_F , and thus on the number of realizations of the random variables \mathbf{x} con-
 188 sidered during the optimization process. This limits the online use of such
 189 an approach through embedded systems.

190 3. Inverse Model for System Identification

191 The inverse model approach presented in this paper is an effective candi-
 192 date for the FC ECM identification [20].

The inverse model denoted as \mathcal{M}^{-1} goes in the opposite direction with
 respect to forward model $\bar{Z}_F(f; \mathbf{x})$ used in the conventional optimization
 approach (2); it is:

$$\mathbf{x}_* = \mathcal{M}^{-1}(\mathbf{Z}_{exp}), \quad (3)$$

193 where, the vector $\mathbf{x}_* \in \mathbb{R}_p^N$ collects the identified system parameters, \mathbf{Z} is
 194 a vector collecting the experimental impedance values evaluated at the fre-
 195 quency $\mathbf{f} = [f_1, \dots, f_{N_f}]^T$ and the inverse map $\mathcal{M}^{-1} : \mathbb{C}^{N_f} \rightarrow \mathbb{R}^d$.

196 Specifically, starting from the experimental impedance values \mathbf{Z}_{exp} , the
 197 inverse map \mathcal{M}^{-1} is able to directly provide, without requiring to solve any
 198 optimization problem or to use an iterative algorithm, the optimal configu-
 199 ration of the ECM parameters \mathbf{x}_* . Indeed, by means of the inverse model,
 200 the identification task becomes as simple as a trivial function evaluation.

201 Unfortunately, for realistic cases in which the inverse map cannot be
 202 computed explicitly from the forward one, the construction of the inverse
 203 model is rather challenging. First of all, even if a mathematical relationship
 204 for the forward model is known, the inverse model might not be available
 205 in a closed-form. In such cases, the model must be constructed numerically
 206 with the help of regression or interpolation techniques, starting from a set of
 207 experimental and/or synthetic training data provided by the forward model.
 208 Moreover, inverse models are often ill-posed, since they can lead to a one-to-
 209 many map (i.e., the inverse model turns out to be a non-injective function),
 210 in the sense that a given set of value of the model output might be generated
 211 by more than one combinations of the system parameters.

212 Advanced ML regression techniques are promising candidates for building
 213 an accurate approximation of the inverse map in a high dimensional space. In
 214 this paper, the effectiveness of three ML regressions, i.e., SVM regression [25],
 215 LS-SVM regression [26] and the GPR [27], is investigated.

216 3.1. SVM and LS-SVM regression

Let us consider the problem of constructing the inverse map \mathcal{M}^{-1} , start-
 ing from a set of N_s training samples $\mathcal{D} = \{(\mathbf{x}_l, \mathbf{y}_l)\}_{l=1}^{N_s}$, in which the vector
 $\mathbf{x}_l \in \mathbb{R}^{N_p}$ collects the training realizations of the system parameters and
 $\mathbf{y}_l = [\text{Re}\{Z_{1,l}\}, \dots, \text{Re}\{Z_{N_f,l}\}, \text{Im}\{Z_{1,l}\}, \dots, \text{Im}\{Z_{N_f,l}\}]^T \in \mathbb{R}^{2N_f}$ is a real
 value vector collecting the corresponding real and imaginary parts of the FC
 impedance values, the latter being synthetic or experimental data. The pri-
 mal space formulation of the inverse model $\tilde{\mathcal{M}}_i^{-1}$ for i -th component of the
 parameter vector \mathbf{x} built via the SVM and LS-SVM regression, writes [25, 26]:

$$x_i \approx \mathcal{M}_i^{-1}(\mathbf{y}) = \langle \mathbf{w}_i, \phi(\mathbf{y}) \rangle + b_i \quad (4)$$

217 where $\phi : \mathbb{R}^{N_p} \rightarrow \mathbb{R}^D$ is a vector collecting D -basis functions ¹, $\mathbf{w}_i \in \mathbb{R}^D$ is a
 218 vector collecting the regression unknowns and b_i is the bias term. The above
 219 formulation can be repeated to model all the components of the vector \mathbf{x} .

¹For the sake of simplicity, it has been assumed that all the components of parameter
 vector x_i share the same basis functions.

For the LS-SVM and SVM regressions, the unknowns (i.e., \mathbf{w}_i and b_i) are estimated by solving the following optimization problem:

$$\min_{\mathbf{w}_i, b_i} \frac{1}{2} \|\mathbf{w}_i\|_{L_2}^2 + \gamma_i \sum_{l=1}^{N_s} \ell(x_{i,l}, \langle \mathbf{w}_i, \phi(\mathbf{y}_l) \rangle + b_i) \quad (5)$$

where $\ell(\cdot)$ is a cost function proving the error of the model on the training samples. This means that the coefficients \mathbf{w}_i and the bias term b_i are estimated by minimizing the average of the cost function evaluated between the training data and the model predictions penalized by a L2 regularizer (i.e., $\|\mathbf{w}_i\|^2$). The latter term adds a penalty to the model accuracy on the training samples, thus mitigating the detrimental effects of the ill-posed problem and also reducing overfitting [34]. The effect of the regularizer in the optimization problem in (5) is tuned by the hyperparameter γ_i , usually estimated via cross-validation (CV) during the model training. A squared loss function is used as cost function for the LS-SVM regression², whereas the SVM regression uses the ε -insensitive loss function. Interested readers can refer to [25] for additional details.

It is important to notice that the primal space formulation of the LS-SVM and SVM regression provides a parametric model, in which the number of unknowns to be estimated (i.e., the cardinality of the vector $|\mathbf{w}| = D$) is equal to the number of basis functions considered in the liner expansion in (4). This means that the obtained model suffers the curse of dimensionality (i.e., a reduction of the model efficiency when either the model complexity and the number of parameters increases).

The dual formulation available for the LS-SVM and SVM regression allows mitigating the above issue and writes:

$$x_i = \sum_{l=1}^{N_s} \alpha_{i,l} k(\mathbf{y}, \mathbf{y}_l) + b_i, \quad (6)$$

where the coefficients $\alpha_{i,l}$ and the bias term b_i are estimated by the regression algorithm during the training process and $k(\cdot, \cdot) : \mathbb{R}^{2N_f \times 2N_f} \rightarrow \mathbb{R}$ is the kernel function defined as the inner product of between the basis function vectors, i.e., $k(\mathbf{y}, \mathbf{y}_l) = \langle \phi(\mathbf{y}), \phi(\mathbf{y}_l) \rangle$. Thanks to the kernel function $k(\cdot, \cdot)$, the dual

²For mathematical convenience, in the LS-SVM regression the squared loss function is multiplied by a constant term 1/2.

243 space formulation in (6) does not require an explicit definition of the basis
 244 functions $\phi(\mathbf{y})$. This is the so-called “kernel trick” [25]. Several kernel func-
 245 tions with different mathematical properties and features are available [35].
 246 The most common ones are:

- 247 • linear kernel: $k(\mathbf{y}, \mathbf{y}_l) = \mathbf{y}^T \mathbf{y}_l$;
- 248 • polynomial kernel of order q : $k(\mathbf{y}, \mathbf{y}_l) = (1 + \mathbf{y}^T \mathbf{y}_l)^q$;
- 249 • Gaussian radial basis function (RBF) kernel: $k(\mathbf{y}, \mathbf{y}_l) = \exp(-\|\mathbf{y} - \mathbf{y}_l\|^2/2\sigma^2)$,
 250 where σ is the kernel hyperparameter.

251 Hereafter, in this paper the Radial Basis Function (RBF) kernel will be
 252 adopted, since it has shown an excellent performance in high nonlinear re-
 253 gression problems [25, 28].

254 In the above dual formulation of the SVM and LS-SVM regression, the
 255 number of regression unknown $\alpha_{i,l}$ turns out to be independent from the di-
 256 mensionality of the input space (i.e., the cardinality of \mathbf{y}). This is extremely
 257 useful to overcome the curse-of-dimensionality issue. Moreover, the regular-
 258 izer used in the primal space formulation allows us to mitigate the ill-posed
 259 problem resulting from the inverse map.

260 It should also be noted that the unknowns of the dual form of the LS-
 261 SVM formulation can be suitably estimated as a solution of a linear problem
 262 by inverting a square matrix [26]. On the other hand, due to the ε -insensitive
 263 loss function, the convex optimization problem for the case of the SVM leads
 264 to a sparse solution, but it must be solved numerically [25].

265 3.2. GPR Regression

Differently from the LS-SVM and SVM regression, the GPR can be used
 to build a probabilistic version of the inverse surrogate model. Starting from
 the N_s training samples $\mathcal{D} = \{(\mathbf{x}_l, \mathbf{y}_l)\}_{l=1}^{N_s}$, the GPR allows to train a prob-
 abilistic inverse model able to estimate for any configuration of the input
 parameters a statistical interpretation of its prediction in terms of a Gaus-
 sian distribution, which provides additional information on its reliability [27].
 Specifically, for any test sample \mathbf{y}_* , the output of a GPR model is:

$$x_{i,*} \sim p(x_{i,*} | \mathbf{y}_*, \mathcal{D}) = \mathcal{N}(\mu_{\mathbf{y}_*}, \sigma_{\mathbf{y}_*}^2) \quad (7)$$

in which $\mu_{\mathbf{y}_*}$ and $\sigma_{\mathbf{y}_*}^2$ are the so-called posterior mean and variance, computed as follows:

$$\mu_{\mathbf{y}_*} = \mathbf{k}_*^T (\mathbf{K} + \sigma_n^2 \mathbf{I})^{-1} \mathbf{x} \quad (8)$$

$$\sigma_{\mathbf{y}_*}^2 = k_{**} - \mathbf{k}_* (\mathbf{K} + \sigma_n^2 \mathbf{I})^{-1} \mathbf{k}_*^T \quad (9)$$

where $\mathbf{y} = [y_1, \dots, y_{2N_f}]$, $\mathbf{K} \in \mathbb{R}^{N_s \times N_s}$ is the correlation matrix in which the covariance function k is evaluated on pairs of the training data (i.e., $K_{ij} = K(\mathbf{y}_i, \mathbf{y}_j)$), $\mathbf{k}_* = [k(\mathbf{y}_1, \mathbf{y}_*), \dots, k(\mathbf{y}_L, \mathbf{y}_*)]^T$ is a column vector, $k(\mathbf{y}_*, \mathbf{y}_*)$ is a scalar, $\mathbf{I} \in \mathbb{R}^{N_s \times N_s}$ is the identity matrix and σ_n^2 is an hyperparameter representing the variance of a possible additive noise corrupting the training dataset. Similarly to the kernel function, the covariance function used in the GPR defines the correlation between the values of pairs of model output at different point in the parameters space. Hereafter in this paper, a squared exponential covariance function, also known as RBF kernel, is used for the GPR, i.e., $k(\mathbf{y}, \mathbf{y}') = \exp(-\|\mathbf{y} - \mathbf{y}'\|^2 / 2\sigma_l^2)$ [27], where σ_l is a hyperparameters estimated during the model training.

4. Learning framework for the inverse model

The LS-SVM, the SVM and the GPR regression models learn the inverse model through a suitable framework. To learn the inverse function of the Fouquet model through a data-driven regression technique, it is necessary to observe both the domain and the co-domain of the function $\mathbf{x} = \mathcal{M}^{-1}(\mathbf{y})$. However, given the very high dimensionality of the two spaces, one depending on the number of frequencies, the other on the number of parameters, an unconstrained generation of observations would be computationally intractable. It is worth to note that, since the forward model $\bar{\mathbf{Z}} = \bar{Z}_F(\mathbf{x}, \mathbf{f})$ is available, the sampling can be limited to the parameters space and the spectrum obtained through the evaluation of the direct model for each sampled parameters' set. However, even in this case, the training procedure would benefit from a bounded search space. From a computational perspective, bounding the parameters values between specific maxima and minima values drastically reduce the training time. Additionally, from a physical perspective, only certain combinations of parameters are feasible and lead to a meaningful impedance spectra, thus only certain regions of the parameters space are worth to be explored.

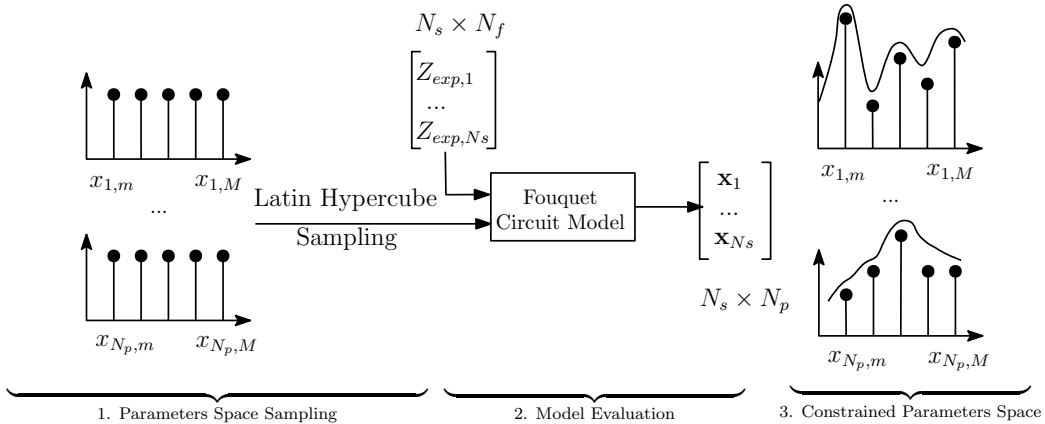


Figure 1: Generation of the training set. Each set of parameters is sampled through the LHS method and is processed with the Fouquet model in the Grid Search Procedure.

295 A synthetic dataset, which is obtained through a specific data-generating
 296 procedure shown in Fig. 1, has been employed for training purposes. It em-
 297 ploys the grid search optimization briefly described in Sec. 2 together with
 298 the Fouquet ECM in which the candidate sets of parameters are sampled
 299 from the search space via a Latin Hypercube Sampling (LHS) scheme. In
 300 the figure, N_s refers to the number of training samples, $N_p = 6$ is the num-
 301 ber of parameters involved in the Fouquet ECM, i.e. the dimension of the
 302 parameters vector \mathbf{x} , and $N_f = 48$ is the number of frequency values.

303 4.1. Parameters Space Sampling

304 Fig. 1 shows that, at the beginning of the procedure, the parameters
 305 space is sampled in the range between \mathbf{x}_m and \mathbf{x}_M . These limits have been
 306 set after having analysed the experimental impedance spectra corresponding
 307 to the extreme fault conditions. Indeed, the experimental data have been
 308 obtained by inducing stack faults with an increasing level of severity. By
 309 moving the stack operating condition away from the nominal one, impedance
 310 modulus and phase change and lead to visible effects on the spectra, e.g. in
 311 terms of a magnification or reduction of the arcs in the Nyquist diagram.
 312 The effect depends on the specific fault, on its severity, and on the duration
 313 of the fault condition. For a specific operating condition imposed at stack
 314 level, the spectra of its cells that are connected in series may not exhibit the
 315 same alteration, because of inhomogeneities in the gas flows, constructive
 316 mismatching among cells and differences in cells degradation rate.

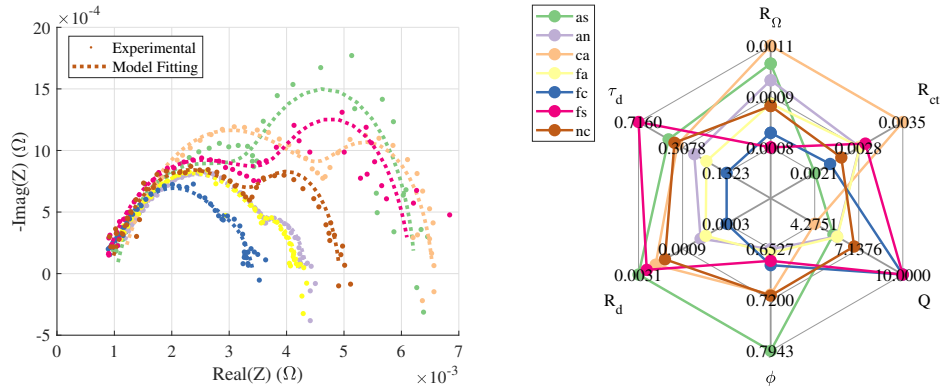


Figure 2: EIS Impedance curves (top panel) in nominal and faulty conditions identified offline and the corresponding parameters (bottom panel) defining the parameters space.

317 Thus, the boundaries for each parameter range have been fixed by picking,
 318 in a subset of curves referring to the same operating condition, the spectrum
 319 that is the farthest from the nominal one. The resulting spectra are consid-
 320 ered as extreme ones and selected for a robust identification. The parametric
 321 identification has been performed according to the approach described in [36].

322 The fluctuation of the experimental measurements around the GA-based
 323 identified impedance curve is shown in Fig. 2. In the figure, the curves refer
 324 to the fitting while the dots are the EIS measurements of the farthest curves
 325 from the reference. Among the selected curves, the extreme values of each
 326 parameter are considered. Fig. 2 also shows the coverage of the parameters
 327 space obtained through the described procedure, together with the obtained
 328 identified parameters set for each specific operating condition. The colors
 329 are used to distinguish the different FC working conditions: Air Starvation
 330 (*as*), Anode and Cathode Drying (*an* and *ca*), Anode and Cathode Flooding
 331 (*fc* and *fa*) Fuel Starvation (*fs*) and Normal Conditions (*nc*).

332 4.2. Grid Search Optimization and the reduced parameters space

333 The parameters space defined in the previous subsection and depicted
 334 in Fig. 1 with the left-most stem plot, is sampled through a LHS proce-
 335 dure and evaluated during the grid search optimization to obtain the set
 336 of solutions $\mathbf{X}_{MC} = [\mathbf{x}_1, \dots, \mathbf{x}_{N_s}]$ for the experimental impedance spectra
 337 $[\mathbf{Z}_{exp,1}, \dots, \mathbf{Z}_{exp,N_s}]$. Thereby, the set of solutions \mathbf{X}_{MC} is used to reduce the
 338 initial parameters space and build a prior discrete N_p -dimensional distribu-
 339 tion of the parameters. This is represented by the rightmost stem plot in

340 Fig. 1. The obtained discrete distribution is smoothed with a Kernel Density
341 Estimate (KDE) procedure so that it can be sampled to obtain an expanded
342 training set of arbitrary dimension [37]. This procedure ensures that the
343 three regression algorithms are focused on the plausible regions of the pa-
344 rameters space, which are those ones leading to impedance spectra that are
345 representative of the FC operating conditions. Moreover, the availability of a
346 continuous distribution from which an arbitrary number of spectra is drawn,
347 allows to benchmark the performance of the regression algorithms by using
348 different sizes of the training set.

349 5. Results on synthetic data

350 The proposed approaches are firstly validated by using the synthetic
351 dataset. Both the training and test sets are sampled from the reduced pa-
352 rameters' space that is obtained as it has been described in Section 4.2. The
353 reduced parameters' space is obtained by using the 30% and 70% of the
354 experimental spectra, in order to create the training and the testing sets
355 respectively. Thus, at this stage, the fractions of the experimental dataset
356 reported above are exploited to estimate the parameters' distributions. The
357 resulting distributions for the training and test sets are then sampled and
358 processed through the Fouquet ECM to obtain the corresponding synthetic
359 impedance spectra that are used for training and test purposes.

360 The use of spectra that do not come from experimental measurements
361 but that have been generated by using the Fouquet ECM allows to analyse
362 the performance of the regression algorithms. Indeed, to each synthetic spec-
363 trum, belonging either to the training set or to the test set, corresponds a set
364 of parameters of the Fouquet ECM that guarantees the perfect fitting of the
365 spectrum thereof. Consequently, modelling inaccuracies of the Fouquet ECM
366 for the available experimental data do not affect the identification result and
367 the performance of the regression approaches can be analysed in detail. The
368 use of synthetic data also allows to study the impact of the number of train-
369 ing samples and of the noise affecting the data in the Nyquist plane, the
370 latter being added through a simple numeric procedure applied to the test
371 set. The noise has been added to the smooth impedance generated through
372 the Fouquet ECM. The spectroscopy measurements are known to be affected
373 by a stochastic noise, whose standard deviation is a function of the frequency
374 and the magnitude of the impedance vector [38]. Therefore, to mimic the

375 noise of the available experimental spectra, the impedance used to build the
 376 synthetic test set has been altered according to (10):

$$Z(f) = \bar{Z}_F(f; \mathbf{x}_*) + \underbrace{(\beta_{re}(f) + j\beta_{im}(f))}_{n(f)}, \quad (10)$$

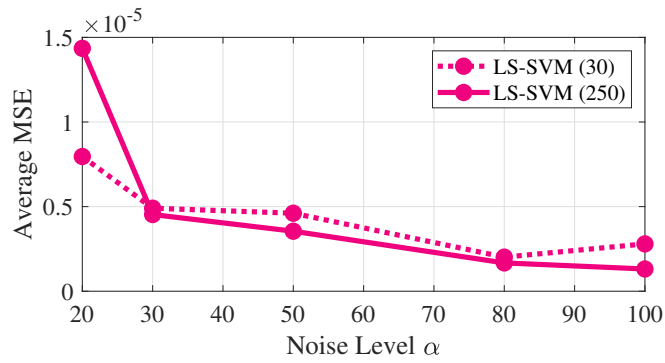
377 where $\beta_{re}(f), \beta_{im}(f) \sim \mathcal{N}(0, \sigma^2(f))$ are Gaussian distributed stochastic vari-
 378 ables describing a non-stationary additive noise $n(f)$, in which the standard
 379 deviation is computed according to (11):

$$\sigma(f) = \frac{|\bar{Z}_F(f; \mathbf{x}_*)|}{\alpha} \quad (11)$$

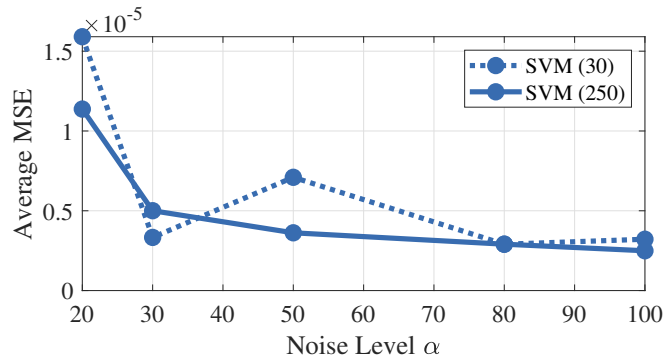
380 and α is used to change the noise level.

381 The amplitude of the noise term $n(f)$ is proportional to the impedance
 382 modulus, and affects its real and imaginary parts independently.

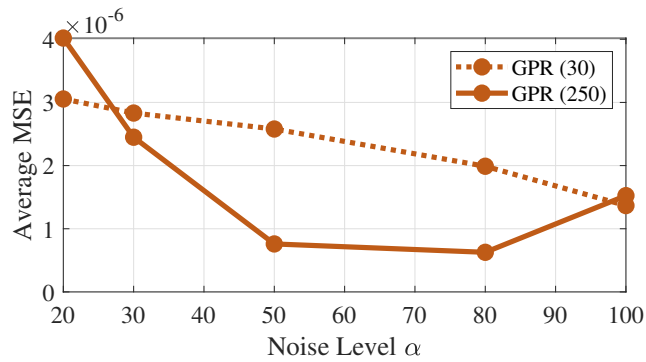
383 Fig. 3 shows the average performance, over three consecutive runs, of
 384 the three algorithms on the test set. The algorithms are trained to infer
 385 the vector of parameters \mathbf{x}_* starting from the impedance Z . The evaluation
 386 of the Fouquet model returns the estimated impedance $\bar{Z}_F(f)(f; \mathbf{x}_*)$. The
 387 average MSE norm between the estimated spectra and the ones of the test
 388 set is used to compare the results. In Fig. 3(a) the LS-SVM performance
 389 underlines the impact of the noise level for two different sizes of the training
 390 set. For high levels of noise, thus α lower than 30, the average performance
 391 with 250 samples is worst than the one obtained with the reduced sample size
 392 of 30. When the noise level decreases, the impact of the increased dataset size
 393 becomes visible. At $\alpha = 100$ the performance index is halved. Similar results
 394 are also obtained for the GPR model (Fig. 3(c)) and very similar results
 395 between the two dataset dimensions have been reached when considering
 396 $\alpha = 100$. It follows that, with the proposed parameters space sampling, the
 397 impact of a reduced training set size is marginal and the size of the training
 398 set becomes important only for low levels of noise.



(a)



(b)



(c)

Figure 3: Influence of the number of training samples and of noise level α on the average MSE computed on the synthetic test set: (a) LS-SVM (b) SVM (c) GPR. Each point is the average of three training runs.

399 *5.1. Hyperparameters optimization*

400 For the SVM and the LS-SVM regressions, the optimization of the model
401 hyperparameters is performed through a 10-fold CV [34] scheme and a Bayesian
402 optimization [39]. Whilst, the GPR hyperparameters are optimized by max-
403 imizing the likelihood on the training set via a grid search approach. The
404 above implementations automate the parameters tuning procedure, thus en-
405 abling the applicability of the methods to different ECMs with small changes,
406 often outperforming the standard hand-tuning approaches. The procedure
407 is executed on the hyperparameters domain defined as it follows. For the
408 SVM and the LS-SVM these are defined in the range $C : (1 \times 10^{-6}, 5 \times 10^4)$,
409 $\gamma : (1 \times 10^{-6}, 5 \times 10^4)$ and $\varepsilon : (1 \times 10^{-11}, 1 \times 10^2)$ (only tuned for the SVM re-
410 gression). For The GPR, the hyperparameters' ranges are $\sigma_l : (1 \times 10^{-5}, 1 \times 10^5)$
411 and $\sigma_n^2 : (1 \times 10^{-5}, 1 \times 10^5)$.

412 **6. Experimental Results**

413 The described procedure has been tested on an experimental dataset [1]
414 described in the Introduction. The parameters space is built by using the
415 procedure described in the previous section and the grid search optimization
416 is employed to narrow the search space operating on 70% of the dataset,
417 corresponding to 110 impedance spectra, while 30% of the set is left for the
418 results evaluation. The histogram plots shown in Fig. 4 give a comprehensive
419 view of the obtained results in terms of the MSE resulting by the comparison
420 of the experimental data with the Fouquet impedance at the same frequency
421 values. The MSE is obtained by using the parameters sets that are inferred
422 by the three regression algorithms on all the spectra of the testing set. Af-
423 ter, for each inferred parameters set, the Fouquet impedance is computed
424 and the MSE is calculated with respect to the corresponding experimental
425 data. Consequently, the MSEs histograms shown in Fig. 4 summarize the
426 performance of the three approaches on all the spectra of the test set. Tab. 1
427 reports the identification results in terms of mean, median, and interquartile
428 range (IQR) of the MSE and MAPE calculated over the spectra belonging to
429 the test set. The mean is reported as an overall performance index and the
430 median is provided to quantify the robustness of the result over the entire
431 test set. The closer the mean and median values the better the identification
432 performance. In this case, the error histogram approaches a normal distri-
433 bution, indicating that there are no reconstructed spectra with an error that

Table 1: Mean, Median, and Interquartile Range (IQR) of the MSE and MAPE computed on the whole test set

	MSE ($\times 10^{-5}$)			MAPE		
	Mean	Median	IQR	Mean	Median	IQR
GPR	0.3179	0.2419	0.2017	0.9985	0.9858	0.0358
SVM	0.3403	0.2325	0.2109	1.0875	1.0844	0.0492
LS-SVM	0.7387	0.5537	0.5407	1.0863	1.0779	0.0438

434 differs significantly from the mean. The interquartile range is also provided
 435 to assess the variability of the obtained result.

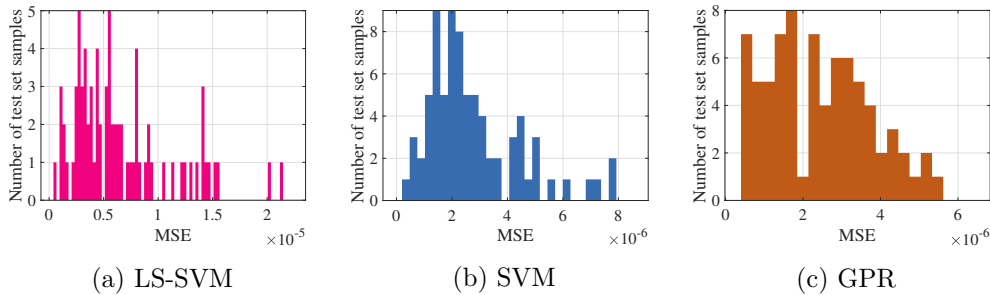


Figure 4: Histogram of the MSE errors computed between the inverse model reconstructions of the impedance spectra and the experimental measurement of the testing set. Each bar corresponds to the number of spectra of the test set giving an MSE falling in the corresponding range.

436 The GPR regression, for a larger number of test set spectra, gives a
 437 smaller error, both in comparison with those ones ensured by the other meth-
 438 ods. Moreover, the reduced spread of the values of the histogram plot indic-
 439 ates that the obtained performance is well generalized on the whole test
 440 set. The SVM shows an average performance similar to the GPR but with
 441 an increased spread of the error histogram. Contrarily, the LS-SVM shows a
 442 performance degradation and higher variability of the result. It is important
 443 to point out that the low performance of the LS-SVM regression could be
 444 due to the quite large range of variation the hyperparameters explored during
 445 the training phase. To quantify the results, different degrees of performance
 446 have been considered.

447 Experimental EIS spectra are affected by an evident noise and artifacts,
 448 thus those ones resulting with an error below 2×10^{-6} are considered as
 449 correctly identified. Fig. 4 shows that GPR and SVM guarantees that the

450 45% and the 41%, respectively, of the spectra in the test set are very well
 451 identified, because the error keeps below the defined threshold. The LS-
 452 SVM gives a 10% of the spectra that are identified with an error due to noise
 453 artifacts. The identification performance is reported in Fig. 5. In both the
 454 cases, the GPR and SVM spectra show optimal results with an MSE below
 455 2×10^{-6} . The results shown in Fig. 5(a) and (b) are obtained through LS-
 456 SVM and are reconstructed with an error equal to 2.8×10^{-6} and 1.5×10^{-5} ,
 457 respectively.

458 Also, the range $2 \times 10^{-6} < \text{MSE} < 2.5 \times 10^{-6}$ is giving valuable solutions.
 459 For the GPR, SVM and LS-SVM models, respectively 11% and 14% and 4%
 460 of the test set are reconstructed with this error. An example of the quality
 461 of the reconstruction obtained in this range is shown in Fig. 6.

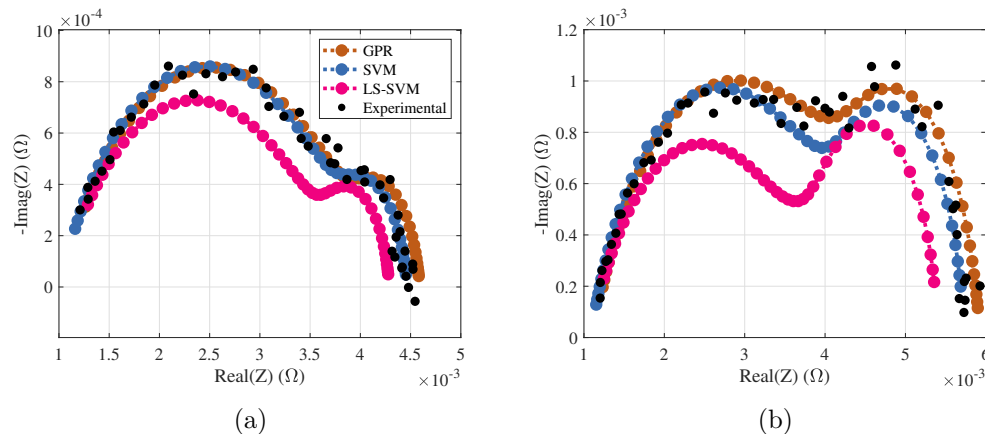


Figure 5: Best identification performance obtained by the GPR and SVM inverse models.

462 7. On Field Results

463 An important point of strength of the proposed inverse model is the short
 464 execution time observed during the phase of inference. Although the training
 465 is computationally expensive due to the dimensionality of the space to be ex-
 466 plored and due to the automatic hyperparameters tuning, the trained model
 467 consists of a simple function, whose evaluation on the measured impedance
 468 spectra returns the set of parameters. For this reason, differently from other
 469 standard model identification procedures relying on iterative algorithms, the
 470 inference through the inverse model is a good candidate for the implemen-
 471 tation on a low cost embedded system, since the impact on the resources

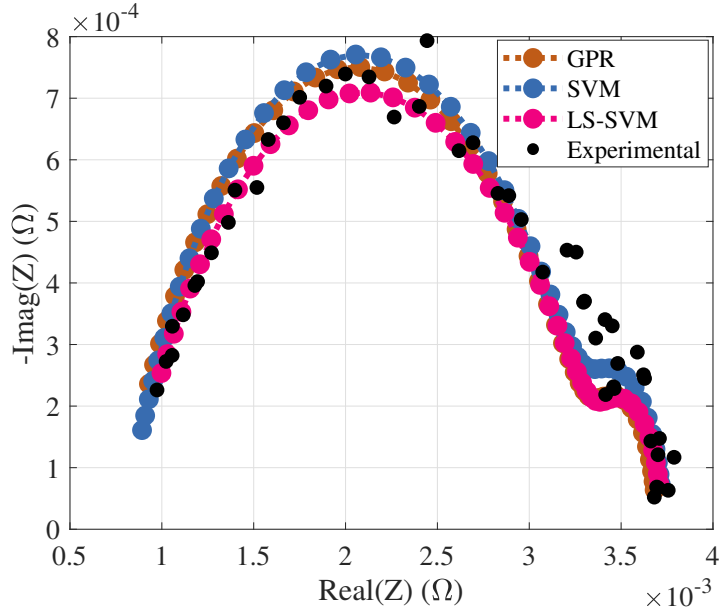


Figure 6: Middle quality identification obtained by the three inverse models.

472 usage is negligible and the execution time is deterministic. Both the latter
 473 aspects are in favor of the approach proposed in this paper with respect
 474 to any stochastic optimization procedure for parameters identification
 475 [22, 23, 40, 41, 42]. The three algorithms are implemented in Python
 476 and executed in a decentralized way. The training procedure, that is compu-
 477 tationally expensive due to the hyperparameters optimization procedure, is
 478 executed on the cloud by employing the Google Colab platform. The remote
 479 machine is equipped with a Dual-Core Haswell Intel Xeon processor at 2.30
 480 GHz and 12 GB of RAM. After the training procedure, the testing is exe-
 481 cuted on a Raspberry Pi 3 embedded system equipped with a Quad-Core 1.2
 482 GHz Broadcom BCM2837 CPU and 1 GB of RAM and the execution times
 483 of the three algorithms have been considered as benchmark. In Fig. 7(a),
 484 the training time of the three models is reported as a function of the size
 485 of the training set. The training time is not only affected by the number
 486 of samples in the training set, but also by the number of hyperparameters
 487 to be tuned and the optimization scheme. This explains the scale difference
 488 between the three techniques. In fact, the training of the SVM regression
 489 turns out to be the most expensive one, since it requires the tuning of three

490 hyperparameters via the CV. On the other hand, the training is faster for
 491 the GPR because the optimization of the hyperparameter is carried out on
 492 the training samples directly, without requiring to compute the CV error.
 Fig. 7(b) reports the execution time for the inference of a set of parameters.

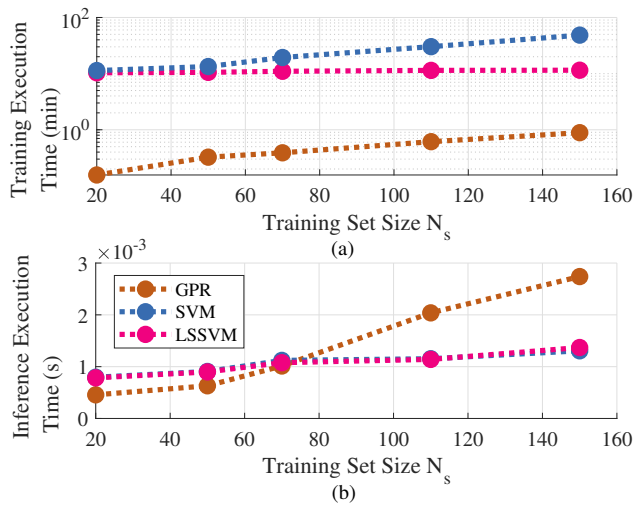


Figure 7: Training (a) and inference (b) execution times of the three regression algorithms.

493
 494 The results clearly highlight the impact of the dimensionality of the training
 495 set on the inference time. Indeed, for the considered approaches, the model
 496 complexity increases with the number of training samples considered during
 497 the model training. The three algorithms infer the parameters set in few
 498 milliseconds. The chosen regressions trained with 110 samples perform the
 499 inference in 2 milliseconds for GPR and in slightly above 1 millisecond for
 500 the SVM and the LS-SVM models, respectively.

501 It is worth to have a comparison in terms of computation time also with
 502 approaches based on parameters identification from EIS data through the
 503 minimization of an objective function value [22][23][43][40]. The objective
 504 function is computed by fixing a set of Fouquet ECM parameters and a set
 505 of frequencies, then evaluating the Fouquet impedance and computing the
 506 MSE between the generated impedance and the experimental data. This
 507 computation has been performed in Python, like the other inverse models
 508 solutions, and run on the same Raspberry Pi 3 mentioned above. The average
 509 execution time is equal to 0.7 ms. This means that a competitive optimization

510 algorithm should be able to find the solution in less than 3 iterations to have
 511 an execution time that is comparable with that one of the GPR inference
 512 time, which is slightly above 2ms.

513 8. Conclusions

514 Machine learning regression models demonstrated to be an efficient choice
 515 to solve the inverse problem with minimal and deterministic execution time
 516 reported during inference. Leveraging the a priori knowledge on the ranges
 517 of the model parameters as well as the knowledge on the regions of the pa-
 518 rameters space that are more likely to correspond to actual impedance mea-
 519 surements, the problem becomes computationally treatable with a training
 520 time that is lower than one minute for the GPR.

521 The three techniques, especially SVM and LS-SVM, have shown to be
 522 more sensitive to noise than to the dataset size. Moreover, for very high
 523 levels of noise, a big data set size has shown to be counterproductive. The
 524 LS-SVM showed weaker results in terms of reconstruction error due to its
 525 sensitivity to the hyperparameters' tuning and execution times similar to
 526 the SVM. Therefore it can be considered a valuable option when a small and
 527 noisy training set is available. Indeed, in the latter case the poor performance
 528 of the three techniques are comparable and the simplicity of the method is
 529 better exploited. The GPR and the SVM have shown very promising results
 530 both in simulated and experimental environment. GPR has been able to
 531 give an optimal reconstruction of the 45% of the spectra and a moderately
 532 good reconstruction of a further 11%. SVR gives a slightly lower accuracy
 533 that is compensated by the execution time required for the inference of the
 534 parameters, which is almost halved with respect to the execution time of the
 535 GPR.

536 Appendix A. Fouquet Equivalent Circuit Model

537 The Fouquet equivalent circuital model has already been adopted in
 538 [21][36][2] for the identification of PEMFC operating in faulty conditions,
 539 and it is defined as follows.

$$Z(f) = \bar{Z}_F(f; \mathbf{x}) = R_\Omega + \frac{\dot{Z}_{\text{CPE}}(\omega)(R_{\text{ct}} + \dot{Z}_{\text{W}}(\omega))}{\dot{Z}_{\text{CPE}}(\omega) + R_{\text{ct}} + \dot{Z}_{\text{W}}(\omega)}, \quad (\text{A.1})$$

540 with

$$\dot{Z}_{CPE}(f) = \frac{1}{Q(j\omega)^\phi}. \quad (\text{A.2})$$

$$\dot{Z}_W(f) = R_d \frac{\tanh \sqrt{j\omega\tau_d}}{\sqrt{j\omega\tau_d}}, \quad (\text{A.3})$$

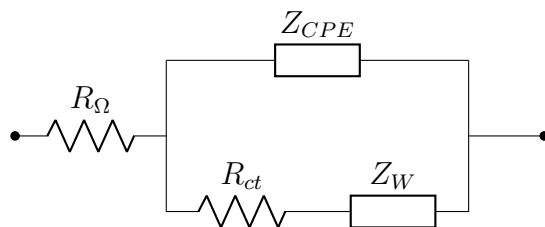


Figure A.8: The Fouquet equivalent circuit model.

541 Here $\omega = 2\pi f$ is the angular frequency. The equivalent circuit model
 542 makes use of a vector of $N_p = 6$ parameters $\mathbf{x} = [R_\Omega, R_{ct}, Q, \phi, R_d, \tau_d]^T$.

543 R_Ω takes into account the losses due to the resistance of the electrolyte to
 544 the flow of protons, while R_{ct} represents the resistance at the electrode/electrolyte
 545 interface to the flow of charges. Q and ϕ are the parameter of the Constant
 546 Phase Element (CPE), used to model the frequency behaviour of the elec-
 547 trodes with rough or porous surfaces. The Warburg element is defined by
 548 R_d , that models the losses due to reactants' diffusion. The time constant of
 549 the diffusion process is specified with the parameter τ_d . The model is evalu-
 550 ated at the same frequencies where the EIS experiment was performed and
 551 $N_f = 48$ impedance points have been obtained.

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