Observations on the Parameter Estimation Problem of Polymer Electrolyte Membrane Fuel Cell Polarization Curves

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Abstract

The optimal operation of fuel cells in changing environmental and variable load conditions requires mathematical modeling. The electrochemical behavior of polymer electrolyte membrane fuel cells (PEMFC) is commonly described with a semi-empirical model requiring fuel cell specific model parameter values. A large number of different nature inspired, heuristic optimization methods have been proposed for this PEMFC parameter estimation problem. In this study, those studies are listed and critically reviewed. In particular, the aim is to elaborate the generalization ability of the results and discuss the fair comparison of the algorithms used for the parameter estimation of the polarization curve. The observations made in this review could further increase the quality of future contributions in this particular area, as well as applications of heuristic optimization methods in other related problems in fuel cell systems.

Keywords: Model Identification, Optimization, Parameter Estimation, Polymer Electrolyte Membrane Fuel Cells, Validation

1 Introduction

Fuel cell technology has shown great promise for partly solving the issue of resource efficiency and clean energy production [1]. It offers a flexible solution for energy conversion with high power density suitable for applications on different scales [2]. Combined with a fuel processing system, hydrogen fuel cells can also be operated with different fuels, such as alcohols, methane, and other hydrocarbons available from various industrial and non-industrial side streams.

Besides the efficiency of fuel cell systems, it is equally important to be able to preserve high performance in changing environmental and variable load conditions by optimizing the operation of the fuel cell. Numerical modeling allows for the investigation of the effect of system parameters, such as different configurations and operating conditions [3]. In 2011, a review by Shah et al. [4] provided a comprehensive introduction to the modeling of polymer electrolyte membrane fuel cell (PEMFC) in different modeling levels. Fuel cell models can be categorized, based on the number of physical dimensions they include (from zero-dimensional to three-dimensional), or by their theoretical and empirical basis (from mechanistic to data-driven) [5]. The zero-dimensional models are simplified, lumped representations which are especially useful in studying the behavior and interactions of fuel cells in power systems.

Electrochemical behavior lies in the heart of most PEMFC models. Typically, this behavior is described by a semi-empirical, zero-dimensional equivalent circuit (polarization curve) model. The earliest representations relied on thermodynamic potential and a simplified loss term [6]. More often than not, the model is based on the generalized steady-state electrochemical model (GSSEM) by Mann, Amphlett et al. [7], where the loss terms of the equivalent circuit model were broken down to have physical meaning. This model has been updated in numerous literature sources with an additional loss term [8–10]. It can also be extended to handle PEMFC aging [11], one of the biggest problems in the long-term operation of PEMFCs [12].

During the past ten years, the parameter estimation of the electrochemical model has attracted a considerable amount of attention. Specifically, it has claimed to offer an industrially relevant example of a nonlinear, complex optimization task. The optimization problem in this case is essentially to find the best parameter combination so that the model would accu-
rately describe the empirically collected PEMFC data. This model can then be used in further power system analysis [13]. In particular, heuristic search methods (evolutionary optimization), such as genetic algorithms (GA), differential evolution (DE) and particle swarm optimization (PSO) with different variants have been applied to the parameter estimation problem. In addition to the above-mentioned optimization problem, different search methods have been successfully applied in the PEMFC stack configuration problem [14, 15], where the aim is to find the optimal PEMFC stack design. However, this study focuses on the former optimization problem.

Many of the studies in this field focus on applying the proposed optimization algorithm solely to the PEMFC model, whereas some of the works contribute to testing the developed algorithms with benchmark functions, as well. The latter can be considered vital, in order to evaluate and generalize the performance of any novel optimization algorithm. However, an optimized PEMFC model also needs to be carefully validated, in order to allow the parameters to represent the fuel cell behavior in varying operating conditions. This part of validation is unfortunately neglected in many cases, making both the generalization of the results and the performance evaluation of the optimization method questionable. Additionally, most of the published research use small data sets for parameter optimization as the literature does not offer rich data sets. This leads to situations where the optimization problem can be anything but challenging.

Recently, two reviews related to this field have been published: (i) Bhatt et al. [16] have reviewed the topic, focusing on GA applications in PEMFC parameter optimization and (ii) Priya et al. [17] have provided a comprehensive review presenting the different optimization algorithms, discussing their performance in PEMFC problems, and analyzing the complexity of the algorithms. In addition to the valuable comments made in those reviews, this study aims further to extend the discussion to the generalization ability of the results, a fair comparison of the algorithms, and offer further critical observations especially regarding the parameter estimation of the PEMFC polarization curve model.

This review is structured as follows: Section 2 presents the most common interpretation of the PEMFC electrochemical model used in parameter estimation problems. In Section 3, the approaches to parameter estimation are collected and compared in terms of numerous features. Section 4 includes the main findings and discussion of this review from several perspectives. Finally, the observations made in this work and future directions for the PEMFC parameter estimation problem are given in Section 5.

2 PEMFC Electrochemical Model

The commonly used representation of the electrochemical model of a PEMFC, showing the unknown model parameters relevant for the PEMFC parameter estimation, is given in Eqs. (2)–(7). The model consists of an expression for a single cell voltage $V_{FC}$, given in Eq. (1), which can be broken down into the internal potential (Eq. (2)), the lumped anode and cathode overvoltage $\eta_{act}$ (Eq. (3)), and the ohmic overvoltage $\eta_{ohm}$ (Eq. (4)). Most of the studies treating the parameter estimation problem also include an additional curve fitting term in the GSSEM known as the concentration overvoltage $\eta_{conc}$ given in Eq. (5). The output voltage of serially connected cells in a PEMFC stack is given in Eq. (6). Hence, the governing equations describe the cell voltage as a function of the cell current $i_{FC}$, limiting current $i_{max}$, and the operating conditions (temperature $T$, hydrogen partial pressure in the anode $p_{H_2}$, oxygen partial pressure $p_{O_2}$, and concentration in the cathode $C_{O_2}$).

$$V_{FC} = E_0 - \eta_{act} - \eta_{ohm} - \eta_{conc}$$ (1)

$$E_0 = 1.229 - 0.85 \cdot 10^{-3}(T - 298.15) + 4.3085 \cdot 10^{-5}T \ln (p_{H_2}F_{O_2})^{0.5}$$ (2)

$$\eta_{act} = -[\xi_1 + \xi_2T + \xi_3T \ln (C_{O_2}) + \xi_4 \ln (i_{FC})]$$ (3)

$$\eta_{ohm} = i_{FC} \left( \frac{R_c + r_m l}{A} \right)$$ (4)

$$\eta_{conc} = -b \ln \left( 1 - \frac{i_{FC} A}{i_{max}} \right)$$ (5)

$$V_{Stack} = N_{Cells} V_{FC}$$ (6)

Naturally, the presented semi-empirical model comprises many simplifying assumptions. They are more thoroughly presented, e.g., in [5, 7]. It has been shown that the model parameters $\xi_1, \xi_2, \xi_3$, and $\xi_4$ can be related to kinetic, thermodynamic and electrochemical phenomena [7]. The partial pressures are typically assumed to be known or expressed as a function of the inlet pressure and relative humidity of vapor in the cathode and anode, respectively, the saturation pressure of the water vapor, cell current, effective electrode area and temperature, as described in [18]. The oxygen concentration can be calculated by Henry’s law once the temperature and oxygen partial pressure are known. The ohmic overvoltage presented in Eq. (4) is an empirical linear correlation of resistances to proton flow as a function of cell current. In Eq. (4), the parameter $R_c$ describes the effect of the electronic resistances and the proton resistances are captured by the fractional term as a function of membrane specific resistivity $r_m$, membrane thickness $l$, and cell active area $A$. Typically, Nafion type membranes are considered and Eq. (7) is used to describe the $r_m$ as a function of $T$ and $i_{FC}$.

$$r_m = \frac{181.6 \cdot \left[ 1 + 0.03(i_{FC} A) - 0.062(T / 303)^2(i_{FC} A)^{2.5} \right]}{[1 - 0.634 - 3(i_{FC} A)] \cdot \exp [4.18(T - 303 / T)]}$$ (7)

In Eq. (7), $\lambda$ is the (bounded) fitting parameter. Finally, the parameter $b$ in Eq. (5) is used to adjust the effect of the mass transport limitations in the high current region. Hence, the
model consists of seven unknown (adjustable) parameters: \( \xi_1 \), \( \xi_2 \), \( \xi_3 \), \( \xi_4 \), \( R_c \), \( L \), and \( b \). This model comprising the GSSEM and the concentration overvoltage is treated here as the nominal model structure, as it appears in 70% of the studies reviewed (see Table 1). It is worth mentioning that the maximum current density \( J_{\text{max}} \) and the permanent current density \( J_n \) associated to the concentration overvoltage term (Eq. (5)) are often used with this model as well (see, e.g., [9]).

### 3 Review of Parameter Optimization for the Electrochemical Model

As mentioned above, parameter estimation of the electrochemical model (and its variants) has been used as an industrially relevant example of testing optimization algorithms for a nonlinear, multivariable, coupled system. Especially since the contributions by Mo et al. [18] and Ohenoja and Leiviska [19], dozens of scientific papers dealing with this problem have been published. Most of this work is listed in Table 1. It should be noted, that although Table 1 contains 45 references, there are still a few more publications on the topic which the

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**Table 1** Reviewed references listed with the data sets and number of data points studied.

| Reference                  | Model structure unaltered | Number of parameters | Search range | Data sets | Number of data points | Different operating conditions | Ratio \( n_s/n_p \) | Notes |
|----------------------------|----------------------------|----------------------|--------------|-----------|-----------------------|-------------------------------|-------------------|-------|
| Mo et al. 2006 [18]        | Yes                        | 7                    | [18]         | Ballard, BCS, SR-12 | 60                     | Yes                           | 8,57             | *Search range established based on several literature references. |
| Ohenoja and Leiviská 2009 [31] | Yes                       | 7                    | [19]         | Ballard, BCS, SR-12 | 10                     | No                            | 1,43             | Simple linear parameter model structure tested as well. |
| Ohenoja and Leiviská 2010 [19] | Yes                       | 7                    | [18], [19]   | Ballard, BCS, SR-12 | 60                     | Yes                           | 8,57             | *Search range expanded based on literature references. Different search ranges tested. |
| Askarzadeh and Rezazadeh 2011a [32] | No*                       | 9                    | [19], [32]** | Ballard     | 14                     | No                            | 1,56             | *Another set of results with 8 parameters \( L_{\text{max}} \) as additional parameter. **Search range with new parameters established. |
| Askarzadeh and Rezazadeh 2011b [69] | Yes/No*                   | 7                    | [18]         | Ballard, BCS, SR-12 | 60                     | Yes                           | 7,5–8,57        | *Another set of results with 10 parameters \( A, l, L_{\text{max}} \) as additional parameters. **Search range with new parameters arbitrarily chosen. |
| Askarzadeh and Rezazadeh 2011c [39] | Yes/No*                   | 7                    | [18], other**| Ballard, BCS, SR-12 | 60                     | Yes                           | 6–8,57          | *Another set of results with 9 parameters \( A, l, L_{\text{max}} \) as additional parameters. **Search range with new parameters established. |
| Askarzadeh and Rezazadeh 2011d [34] | Yes/No*                   | 7                    | [18], [34]** | Ballard, BCS, SR-12 | 60                     | Yes                           | 6,67–8,57       | *Another set of results with 10 parameters \( A, l, L_{\text{max}} \) as additional parameters. **Search range with new parameters established. |
| Askarzadeh and Rezazadeh 2012 [58] | No*                       | 9                    | [32]         | Ballard, BCS, SR-12 | 7–37                   | No                            | 0,78–4,11        | *\( J_n, L_{\text{max}} \) as additional parameters. |
| Chakraborty et al. 2012 [59] | Yes                       | 7                    | [18], [19]   | other*      | 15                     | No                            | 2,14             | *Data from simulation model with noise component. |
| Karimi et al. 2012 [60]    | No*                       | 9                    | [32]         | Ballard     | 14                     | No                            | 1,56             | *\( J_n, L_{\text{max}} \) as additional parameters. |
| Sorsa et al. 2012 [61]     | Yes                       | 7                    | other*       | Ballard, BCS, SR-12 | 60                     | Yes                           | 8,57             | *Search range arbitrarily chosen. |
| Yang and Wang 2012 [55]    | Yes                       | 7                    | [19]         | Ballard, BCS, SR-12 | 60                     | Yes                           | 8,57             | *Search range limits rounded. |
| Askarzadeh and Rezazadeh 2013 [33] | No*                       | 11                   | [32], [33]** | Ballard     | 14                     | No                            | 1,27             | *\( A, l, J_n, L_{\text{max}} \) as additional parameters. **Search range with new parameters established. |
| Askarzadeh 2013 [40]       | No*                       | 11                   | [33]         | Ballard, BCS, SR-12 | 14–37                  | No                            | 1,27–3,36        | *\( A, l, J_n, L_{\text{max}} \) as additional parameters. Another optimization performed with only the most sensitive parameters. |
| Gong and Cai 2013 [62]     | Yes                       | 7                    | [19]         | Ballard, BCS, SR-12 | 60                     | Yes                           | 2,14             | *Only one data set from 250 W fuel cell used. **Data from simulation model with noise component. |
| Reference                        | Model structure unaltered | Number of parameters | Search range                  | Data sets | Number of data points | Different operating conditions | Ratio $n_x/n_p$ | Notes                                                                 |
|---------------------------------|---------------------------|----------------------|-------------------------------|-----------|-----------------------|--------------------------------|----------------|------------------------------------------------------------------------|
| Zhang and Wang 2013 [47]        | Yes                       | 7                    | [18]                          | 250 W     | 60                    | Yes                            | 8,57           | Different search ranges tested.                                        |
| Zhang et al. 2013 [45]          | Yes                       | 7                    | [18]                          | 250 W     | 60                    | Yes                            | 8,57           | Different search ranges tested.                                        |
| Askarzadeh and Coelho 2014 [41]| No*                       | 11                   | [33]                          | Ballard, SR-12 | 14–37                | No                             | 1.27–3.36       | *A, l, Jn, Jmax as additional parameters.                               |
| Cheng and Zhang 2014 [56]       | Yes/No*                   | 7                    | [18],[32]                     | Ballard, BCS, SR-12, 250 W | 7–60                   | Yes                            | 0.78–8.57       | *Another set of results with 9 parameters (Jmax, Jn as additional parameters). Optimization with noise components tested. |
| Gong and Cai 2014 [70]          | No*                       | 10                   | [33]**                        | Ballard, BCS, SR-12, Temasek, other** | 7–50                   | No                             | 0.7–5           | *Jn, Jmax, l as additional parameters. **Part of the data from a simulation model. |
| Niu et al. 2014a [50]           | Yes                       | 7                    | [18], [19]*                   | 250 W     | 60                    | Yes                            | 8,57           | Different search ranges tested. *3rd search range rounded from [19].   |
| Niu et al. 2014b [49]           | Yes                       | 7                    | [18], [19]                    | 250 W     | 60                    | Yes                            | 8,57           | Different search ranges tested.                                       |
| Zhu et al. 2014 [44]            | Yes                       | 7                    | [18], [19]                    | 250 W     | 60                    | Yes                            | 8,57           | Different search ranges tested.                                       |
| Gong et al. 2015 [42]           | No*                       | 11                   | [33]                          | Ballard, BCS, SR-12, Temasek, other** | 7–148                     | No                              | 0.64–13.45      | *Jn, l, A as additional parameters. **Part of the data from simulation model. |
| Lv & Zhang 2015 [48]            | Yes                       | 7                    | [18], [19]                    | 250 W     | 60                    | Yes                            | 8,57           | Different search ranges tested.                                       |
| Sun et al. 2015 [51]            | Yes                       | 7                    | [18], [19]                    | 250 W     | 60                    | Yes                            | 8,57           | Different search ranges tested.                                       |
| Geem and Noh 2016 [9]           | No*                       | 9                    | [32]                          | other     | 13**                  | No                             | 1.44           | *|Jn, l, A as additional parameters. **Search range modified.          |
| Turgut and Coban 2016 [57]      | Yes/No*                   | 7                    | [19], [33]**                  | 250 W     | 60                    | Yes                            | 6–8,57         | *Another set of results with 10 parameters (A, l and Jmax as additional parameters). **Search range modified. *3 additional parameters for $\eta_{unc}$ term. **Search range modified. |
| Yang et al. 2016 [10]           | No*                       | 10                   | [19]**                        | 250 W     | 60                    | Yes                            | 6              | *Search range for $\lambda$ altered. **Only one data set from 250 W fuel cell used. |
| Ali et al. 2017 [71]            | Yes                       | 7                    | [19]                          | Ballard, BCS, SR-12, Temasek | 7–18                   | Yes                            | 1–2.57         | *Search range modified.                                                 |
| El-Fergany 2017 [72]            | Yes                       | 7                    | [19]*                         | 250 W**, Ballard, SR-12 | 13–20             | No                             | 1.86–2.86      | *Calculated value for $\varepsilon_z$ used. **Search range for $\lambda$ altered. Tabulated data set for NedStack PEMFC. |
| El-Fergany 2018 [36]            | No*                       | 6                    | [19]**                        | BSC, NedStack | 18–29             | No                             | 3–4.83         | *Calculated value for $\varepsilon_z$ used. **Search range for $\lambda$ altered. Tabulated data set for NedStack PEMFC. |
| Fathy and Rezk 2018 [52]        | Yes                       | 7                    | [18], [19]                    | 250 W     | 60                    | Yes                            | 8,57           | Different search ranges tested.                                       |
| Chen and Wang 2019 [46]         | Yes                       | 7                    | [18], [19]                    | 250 W, Ballard, BCS, SR-12 | 7–60             | Yes                            | 1–8.57         | Different search ranges tested.                                       |
| Xu et al. 2019 [53]             | Yes/No*                   | 7                    | [18], [19], [34]             | 250 W     | 60                    | Yes                            | 6.67–8.57      | *Another set of results with 9 parameters (l, Jmax as additional parameters). Different search ranges tested. |
| Kler et al. 2019 [64]           | No*                       | 11                   | [33]                          | Ballard, BCS, SR-12 | 15–54             | No                             | 1.36–4.91      | *Jn, l, A as additional parameters. Data set for Ballard, BCS and SR-12 tabulated. |
In Table 1, the publications are tagged with the number of model parameters used ($n_p$), the parameter search range used, and the number of data points ($n_d$) and data set(s) used. Finally, it is indicated whether the data sets cover different operating conditions and whether the model structure proposed, e.g., in [18, 19], is utilized as it is, or altered. Four different parameter search ranges can be identified: the narrow search range originates from the study by Mo et al. [18], the expanded search range is presented in Ohenoja and Leiviskä [19, 31], and the search ranges comprising additional parameters are given in Askarzadeh and Rezazadeh [32–34]. Additionally, some authors have preferred to use rounded values for the search range bounds. Hence, these are noted in Table 1.

The data sets used in the listed studies mostly rely on the examples found in the literature instead of new experimental data. Hence, the data sets can be categorized into six PEMFC types: SR-12, BCS, Ballard, 250 W, Temasek, and NedStack. The data for the first three fuel cells originate from Correa et al. [8], whereas the 250 W is presented in Mo et al. [18] and Temasek in Jia et al. [35]. The NedStack data are tabulated in El-Fergany 2018 [36]. Only the 250 W data set comprises polarization curves attained in different operating conditions. Typically, the data are interpreted from the graphs given in the original publications. In cases, where other fuel cell data sets are used, a note is given in Table 1.

In Table 2, the publications are tagged with the optimization algorithm and the validation method used. For the validation method, it is indicated whether or not the algorithm performance was tested with benchmark functions. Accordingly, it is indicated whether the optimization results are compared to those achieved with different optimizers, and if this comparison is made directly with the objective function (OF) values reported in other studies (being sensitive to errors in data and optimization ranges) or if the results are re-calculated or re-optimized with the same data sets. Finally, additional notes are given in the last column.

4 Discussion

The discussion is divided into four parts: first, the consistency within the utilized model structures is discussed. Secondly, the amount of data used and risks related to the low number of data are highlighted. This is followed by observations on the internal validation strategies and search ranges.

Table 1. Continued.

| Reference                  | Model structure unaltered | Number of parameters | Search range | Data sets                        | Number of data points | Different operating conditions | Ratio $n_d/n_p$ | Notes                                                                 |
|----------------------------|---------------------------|----------------------|--------------|---------------------------------|-----------------------|-------------------------------|----------------|-----------------------------------------------------------------------|
| Kandidayeni et al. 2019 [65] | Yes                       | 7                    | [19]¹         | BSC, NedStack, other**          | 15–29                 | No                            | 2,14–4,14      | *Search range for $\lambda$ altered. **Data sets for BCS, NedStack and Horizon H-12 FC tabulated. |
| El-Fergany et al. 2019 [13]   | Yes                       | 7                    | n.a. *        | Ballard, SR-12, 250 W, other** | 13–20                 | No***                         | 1,86–2,86      | *Search range not reported. **Data set for Horizon H-12 FC tabulated. **H-12 tested in three different conditions, but model parameters fitted separately for each case. |
| Isa et al. 2019 [73]         | No*                       | 11                   | [33]          | Ballard                          | 14                    | No                            | 1,27           | *$J_\text{max, l, A}$ as additional parameters. |
| Duan et al. 2019 [63]        | Yes                       | 7                    | [33]¹         | Ballard, SR-12, BCS, 250 W, Temasek | 7–60                  | Yes                           | 1–8,57         | *Search range for $\xi_1$, $\xi_2$, b and $R_c$ altered. |
| Fawzi et al. 2019 [74]       | Yes                       | 7                    | [19]¹         | Ballard, BSC, NedStack          | 13–29                 | No                            | 1,86–4,14      | *Search range for $\xi_2$ and $\lambda$ altered. |
| Agwa et al. 2019 [75]        | Yes                       | 7                    | [19]¹         | SR-12, 250 W**                  | 15–20                 | No                            | 2,14–2,86      | *Search range for $\xi_2$ and $\lambda$ altered. **Only one data set from 250 W fuel cell used. |
| Fathy et al. 2020 [54]       | Yes                       | 7                    | [18], [19]    | BCS, SR-12, 250 W, NedStack     | 18–60                 | Yes                           | 2,57–8,57      | Different search ranges tested. |
| Menesy et al. 2020a [76]      | Yes                       | 7                    | [19]¹         | BCS, SR-12, 250 W, NedStack     | 15–18                 | No                            | 2,14–2,57      | *Search range for $\xi_2$ and $\lambda$ altered. **Only one data set from 250 W fuel cell used. Data set for 250 W tabulated. |
| Menesy et al. 2020b [66]      | Yes                       | 7                    | [19]¹         | BCS, SR-12, 250 W**, Temasek     | 15–18                 | No                            | 2,14–2,57      | *Search range for $\xi_2$ and $\lambda$ altered. **Only one data set from 250 W fuel cell used. Data set for BSC and 250 W tabulated. |
Table 2: Reviewed references listed with the optimization method and validation strategies applied.

| Ref. | Optimization method                                | Abbreviation | Benchmarking | Different operating conditions | OF comparison | With re-simulated results | Notes                       |
|------|----------------------------------------------------|--------------|--------------|---------------------------------|---------------|--------------------------|-----------------------------|
| [18] | Hybrid genetic algorithm                          | HGA          | No           | Yes                             | Yes           | Yes                      | OF comparison to simple GA. |
| [31] | Genetic algorithm                                  | GA           | No           | No                              | No            | No                       | Statistical performance studied. |
| [19] | Genetic algorithm                                  | GA           | No           | Yes                             | Yes           | No                       | Direct OF comparison to HGA. Statistical performance and validation strategies studied. |
| [32] | Grouping-based global harmony search              | GGH S        | No           | No                              | Yes           | Yes                      | Performance compared to 6 other methods. Statistical performance studied. |
| [69] | Artificial bee swarm optimization                 | ABSO         | No           | Yes                             | Yes           | Yes/No                   | Direct OF comparison to 2 methods in literature. Comparison with optimization results for PSO. |
| [39] | Artificial immune system                          | AIS          | No           | Yes                             | Yes           | Yes/No                   | Direct OF comparison to 2 other methods in literature. Comparison with optimization results for PSO. |
| [34] | Modified particle swarm optimization               | MPSO         | No           | Yes                             | Yes           | Yes/No                   | Direct OF comparison to HGA. Comparison with optimization results for PSO. |
| [58] | Innovative global harmony search                  | IGHS         | No           | No                              | Yes           | Yes                      | Performance compared to 7 other methods. Statistical performance studied. |
| [59] | Differential evolution                             | DE           | No           | No                              | Yes           | Yes                      | DE and GA variants tested. Statistical performance studied. |
| [60] | Tournament selection based harmony search         | TSHS         | No           | No                              | Yes           | Yes                      | Performance compared to 4 other methods. Statistical performance studied. |
| [61] | Differential evolution                             | DE           | No           | Yes                             | Yes           | Yes                      | DE variants tested. Statistical performance studied. |
| [55] | Bio-inspired P systems based optimization         | BIPOA        | Yes          | Yes                             | Yes           | No                       | OF comparison to 3 other methods by re-calculating the OF with parameter values found in literature. |
| [33] | Bird mating optimizer                             | BMO          | No           | No                              | Yes           | Yes                      | Performance compared to 3 other methods. Statistical performance studied. |
| [40] | Bird mating optimizer                             | BMO          | No           | No                              | Yes           | Yes                      | BMO performance compared to 9 other methods, statistical performance studied |
| [62] | Differential evolution                             | DE, rank-DEGL | No           | No                              | Yes           | Yes/No                   | Performance compared to different DE variants and 4 other methods using simulated data. OF comparison to 6 other methods by re-calculating the OF found in literature. |
| [47] | Adaptive RNA genetic algorithm                    | ARNAGA       | Yes          | Yes                             | Yes           | No                       | Direct OF comparison to 3 other methods in literature. |
| [45] | Hybrid artificial bee colony algorithm             | HABC         | Yes          | Yes                             | Yes           | No                       | Direct OF comparison to 3 other methods in literature. Validation strategies tested. |
| [41] | Backtracking search algorithm combined with Burger's chaotic map | BSABCM       | No           | No                              | Yes           | Yes                      | Performance compared to 2 BSABCM variants and 2 other methods. Statistical performance studied. |
| [56] | Adaptive differential evolution                    | ADE          | Yes          | No                              | Yes           | Yes                      | Performance compared to 8 other methods. Statistical performance studied. ADE tested with noised data. |
| [70] | Multi-strategy adaptive differential evolution     | DE, rank-MADE | No           | No                              | Yes           | Yes                      | Performance compared to different DE variants and 4 other methods. |
| [50] | Simplified teaching-learning based optimization algorithm | STLBO | Yes          | Yes                             | Yes           | Yes                      | Performance compared to 5 other methods. |
| [49] | Biogeography-based optimization with mutation strategies | BBOM       | Yes          | Yes                             | Yes           | Yes                      | Performance compared to 5 other methods. |
| [44] | Circular genetic operators based RNA genetic algorithm | cRNAGA    | Yes          | Yes                             | Yes           | No                       | Direct OF comparison to 3 other methods in literature. Validation strategies studied. |
| [42] | Transferred adaptive differential evolution        | DE, TRADE    | No           | No                              | Yes           | Yes                      | Performance compared to different DE variants and 4 other methods. |
Table 2. Continued.

| Ref. | Optimization method | Abbreviation | Benchmarking | Different operating conditions | OF comparison | With re-simulated results | Notes |
|------|---------------------|--------------|--------------|---------------------------------|---------------|--------------------------|-------|
| [48] | Modified multi-group DNA genetic algorithm | MMDNAGA | No | Yes | Yes | No | Direct OF comparison to 3 other methods in literature. |
| [51] | Hybrid adaptive differential evolution | HADE | Yes | Yes | Yes | No | Direct OF comparison to 3 other methods in literature. |
| [9] | Generalized reduced gradient technique | GRG | No | No | Yes | No | OF comparison to 4 other methods by re-calculating the OF with parameter values found in literature. However, those parameters are for a different PEMFC than the ones optimized with GRG. |
| [57] | Teaching learning based optimization method hybridized with Differential evolution | TLBODE | Yes | Yes | Yes | Yes/No | Performance compared to 8 other methods. Statistical performance studied. Direct OF comparison to 4 other methods in literature. With the 10 parameter model, direct OF comparison to 1 other method in literature, and 6 other methods with statistical performance as well. |
| [10] | Aging and challenging P systems based optimization | ACPOA | Yes | Yes | Yes | Yes/No | Performance comparison to SGA. OF comparison to other methods by re-calculating the OF with parameters found in literature. However, those parameters are optimized for different model structures than the ones optimized with ACPOA. |
| [71] | Gray wolf optimizer | GWO | No | Yes | Yes | Yes/No | Performance compared to 2 other methods. Statistical performance studied. Direct OF comparison to 8 other methods in literature. |
| [72] | Grasshopper optimization algorithm | GSO | No | No | Yes | No | Direct OF comparison to 1–3 other methods in literature. |
| [36] | Salp swarm optimizer | SSQ | No | No | Yes | No | Direct OF comparison to 2 other methods in literature. |
| [52] | Multi-verse optimizer | MVO | No | Yes | Yes | Yes/No | Performance compared to 4 other methods. Direct OF comparison with 5 other methods in literature. |
| [46] | Cuckoo search | CS-EO | Yes | Yes | Yes | No | Performance comparison to 4 other methods for benchmarking and statistical performance. Direct OF comparison to 4 other methods in literature. |
| [53] | Two-stage eagle strategy | JAYA-NM | No | Yes | Yes | Yes | Performance compared to 5 other methods. Statistical performance studied. Random data splitting. |
| [64] | Hybrid interior search algorithm | HISA | No | No | Yes | Yes | Performance compared to 7 other methods. Statistical performance studied. |
| [65] | Shuffled frog-leaping, Firefly optimization, Imperialist competitive | SFLA, FOA, ICA | No | No | Yes | Yes | Performance of 3 methods compared. Statistical performance studied. Direct OF comparison to 3 other methods in literature. |
| [13] | Whale optimization algorithm | WOA | No | No | Yes | Yes/No | Statistical performance studied. Comparison with optimization results for GA for one FC. Direct OF comparison to large number of other methods in literature. |
| [73] | Ant lion optimizer and Dragonfly algorithm | ALO, DA | No | No | Yes | Yes/No | Performance compared to one other method. Direct OF comparison to four other methods in literature. |
| [63] | Satin bowlerbird optimizer | SBO | No | Yes | Yes | Yes/No | Performance compared to 3 other methods. Statistical performance studied. Direct OF comparison to 8 other methods in literature. |
| [74] | Neural network algorithm | NNA | No | No | Yes | No | Direct OF comparison to 4–7 other methods in literature. |
| [75] | Atom search optimizer | ASO | No | No | Yes | No | Direct OF comparison to 3–6 other methods in literature. |
Finally, some notes about the fair comparison between the methods are presented.

### 4.1 Model Structure

The original GSSEM is largely mechanistic, having a theoretical basis for most of the terms [7]. The model structure presented in Section 2 is often referred to as a semi-empirical model as it incorporates the above-mentioned GSSEM terms and the concentration overvoltage term, which can be seen as an empirical curve-fitting term. Such empirical terms may have no physical meaning. Attempts to explain the concentration overvoltage term has been made in, e.g., [37, 38]. Nevertheless, the curve-fitting properties of the electrochemical model have been further improved in several studies: For instance, [10] describes the semi-empirical model, but with an additional three extra terms. Based on the extra degree of freedom, this model could also be classified as empirical rather than semi-empirical. In [33, 39–42], the optimization problem is formulated in such a way that the design parameters of the PEMFC \((A, l)\) are used as additional parameters in optimization. This approach loses the connection with the real-life system that was used to capture the data for the modeling since the system had predefined/fixed design parameters. As the model and the optimization problem have been reformulated in many papers, comparison of the results with the other literature is not straightforward. It has been shown that the modeling accuracy of the electrochemical models can also further be improved without additional degrees of freedom if the optimization problem is formulated as a model structure search [43].

### 4.2 Risk of Overfitting

Introducing an optimization problem with a number of data points that is only a little higher than the number of estimated parameters in fact poses a relatively easy optimization problem for any optimization algorithm and is prone to overfitting. With too many input parameters or too few data, the fitted model eventually starts to model the noise present in the data. This results in worse predictions outside the training set. Therefore, the validation of the model with unseen data (testing set) is needed. In the case of PEMFC polarization curves, this step naturally involves utilizing data sets in different operating conditions.

Tables 1 and 2 show that 49% of the papers (22 out of 45) generalize the modeling and parameter estimation problem for different PEMFC operating conditions. In the authors’ point of view, this percentage is low, especially if we take into account the fact that the original GSSEM with one parameter set aimed at describing PEMFC electrochemical behavior under different operating conditions. The risk of overfitting is emphasized in cases where only one data set in a single set of operating conditions is utilized. Thus, the optimization problem only involves the training data set. Indeed, this tendency is shown by comparing the ratio of the number of data points and model parameters \((n_x/n_p)\) shown in Table 1. For the studies utilizing four data sets from a 250 W fuel cell, the optimization problem can be divided into training and testing (validation). This results in a ratio of around 8.6 data points per unknown parameter. For studies using only one polarization curve, the ratio can be less than one and typically does not exceed a value of 4.1, except when simulated data are used.

The possibility of overfitting has not received the attention it deserves in this case. Only a few papers have scratched the surface by presenting different cases of data division into test and validation sets [19, 44, 45]. Most importantly, they highlight that a generalizable set of parameters for different operating conditions cannot be achieved by using only one data set in the optimization. Also, the different data divisions lead to very different parameter values with only small changes in the value of the objective function.

### 4.3 Search Range and Validation Strategy

It has been demonstrated how sensitive the results from an evolutionary optimizer are to changes in the parameter search range and the validation strategy [19]. Most of the studied cases with the 250 W fuel cell start by dividing the data sets so that the validation does not have to extrapolate the results, i.e., the extreme cases are in the training data. The effect of this was also shown in [19] and later in [44, 45]: the different types
of data division (validation strategy) lead to worse results in terms of the objective function. In addition, random data splitting strategies have been applied [46]. Regarding the parameter search space, the optimization results for a constrained problem gained from different search spaces (constraints) make the direct comparison of the algorithm performance impossible. Several studies have demonstrated the sensitivity of the changing parameter search ranges to the optimization result [19, 44–54]. In addition, the careless expansion of parameter ranges in a semi-empirical model can also lead to a result [19, 44–54].

In Table 2 also show that many of the studies have presented the essential comparison of the statistical performance of the optimization algorithms. The statistical measures in these cases contribute on how well the heuristic search method with different initial guesses can reproduce the (global) optimum for a particular optimization task. The performance evaluation for the polarization curve model in many studies is simply based on the direct comparison of the objective function values. Although this can be considered feasible whenever exactly the same benchmark data are available, this is not the case with PEMFC data. Typically, the polarization curves are interpreted from graphical representations, being subject to small errors. Indeed, this topic has been recognized or demonstrated in several studies, see e.g. [49, 50, 53, 55, 56, 62]. A comparison of original and recalculated results has been performed in [10, 55] showing that the objective function value is sensitive to errors in data. In contrast, some of the reviewed studies fail to take this into consideration, and even compare the objective function values gained with different parameter sets (i.e., model structures), parameter search space, and even with different objective functions. Some of the most recent studies have noticed the missing tabulated data sets and have presented the data used [9, 13, 36, 53, 64–66]. Also, the effect of the measurement errors is covered in [56, 59, 62], where a noise component was added to the data. Naturally, the largest changes were seen in the objective function value, with less effect on the parameter values and the polarization curve itself.

With the thresholds utilized in this review (columns 4–7 in Table 2), it can be concluded that only a few studies [49, 50, 56, 57] have been able to generalize both the optimization algorithm performance and its application to PEMFC modeling. These studies compare the different algorithms with benchmark functions, use different operating conditions for the polarization curve model, and finally compare the achieved objective function values with re-simulated results (i.e., the same data sets for optimization) rather than directly using literature values.

Considering the uncertainties related to the data sets, model structures and search ranges, what constitutes a significant improvement in modeling accuracy of PEMFC? Some authors are not shy to conclude that their algorithms are superior to previous work in terms of objective function values. On the other hand, in Askarzadeh [40], the comparison of ten different optimizers for a parameter estimation problem concerning only the most sensitive model parameters revealed that each optimization method performed equally well in terms of the best fit. It should be noted that the optimization was performed on a single polarization curve in this study. There seems to be no discussion on what is the feasible or required modeling accuracy for the electrochemical model in real applications. It is likely that any of the reviewed search algorithms can converge towards the global optimum, even if the objective function is nonlinear and complex. Therefore, the selection of the algorithm in practical situations would more probably be based on criteria, such as computational demand (although not that relevant in an offline application), the complexity of the algorithm, and its implementation, and the number of algorithm-tuning parameters. A more comprehensive analysis of the performance and evaluation of algorithms in terms of complexity, V–I characteristics, objective function, computational speed, and parameter tuning is provided in Priya et al. [17].

5 Conclusions

The most essential observations made in this work regarding the PEMFC parameter estimation problem are related to (i) validating the results in different operating conditions, (ii) the need for more comprehensive data sets, and (iii) the openness of data, source codes and optimization constraints to establish a fair comparison of results. These aspects can hopefully be emphasized in future studies together with an evaluation of the required modeling accuracy on the PEMFC system level. The GGSEM and its variants are primarily targeted to describe the electrochemical performance of PEMFC in vary-
ing operating conditions. Therefore, it is of utmost importance to optimize the parameters with data collected under such circumstances, and as Priya et al. [17] point out, to test the realized models for all operating conditions. Unfortunately, the published PEMFC data are limited in this respect. The data originating from Mo et al. [18] is a starting point, but still only one example with a restricted amount of data points. For a real benchmark test, richer data sets are needed. For instance, the benchmark data could be simulated via a rigorous model in different operating conditions. This would also allow the implementation of (unsupervised) machine learning methods for PEMFC model identification.

With rich PEMFC data sets, the pitfalls of overfitting could also be reduced efficiently. In addition, noise and drift elements could be utilized to stress the optimization algorithm performance, especially in terms of stability and reliability, as emphasized by Xu et al. [53]. In real-time applications, where parameter estimation is performed in an adaptive manner (see, e.g., Ettihir et al. [67], and Xing et al. [68]), the convergence speed would become one of the key performance indicators.

Some papers have presented tabulated data sets [9, 36, 43, 53]. Future studies should preferably be built on these data sets in the absence of mentioned benchmark data. With a consensus on parameter search ranges as well, a more straightforward performance comparison of the parameter estimation problem of the polarization curve could be made. The final observation, also pointed out in [17, 53], is that the availability of codes plays an important role in the fair comparison of optimization methods for the problem of polarization curve parameter estimation.

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