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Nonlinear D-optimal design of experiments for polymer–electrolyte–membrane fuel cells

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ABSTRACT

Using empirical models, parameters have to be estimated from experimental data. Experimental characterization of fuel cell stacks is an expensive and time-consuming task. Therefore it is very important to choose an experimental design, which maximizes the statistical quality of the resulting information. Box and Lucas (Biometrika 46 (1959)) showed that it is possible to optimize nonlinear experimental designs by the minimization of the covariance matrix of the least squares estimate. The aim of this work is to adopt this general method in order to investigate its ability for application in polymer–electrolyte–membrane fuel cell (PEMFC) characterization. Based on an empirical PEMFC model a D-optimal design criterion has been developed and validated. Numerical methods, evolutionary and heuristic are investigated with respect to fast and robust evaluation of the design criterion. For a given set of experimental data best results are achieved using a heuristic approach, a so-called sequential search. Based on that result an algorithm to obtain an optimal design of experiments (DoE) in a nondeterministic operating area is introduced. The proposed algorithm is able to take into account experimental limitations due to test facilities or examinee. The algorithm further allows to include existing and for reference needed experiments.

1. Introduction

Intelligent energy management is a cost effective key path to realize efficient automotive drive trains. To develop operating strategy in fuel cell drive trains, precise and computational efficient models of all system components, especially the fuel cell stack, are needed.

System identification is an essential step in empirical modelling. Especially, the estimation of unknown parameters is a typical problem in the development of a PEMFC model. The standard method is the analysis of experimental data from measurements. Due to system complexity of a PEMFC the experimental investigation is an expensive and time-consuming task. In most of the cases just a limited operating area is available for the measurements caused by the fuel cell capabilities, physical laws, test equipment, or interactions between these components. To give a simple example for equipment limitations just consider the gas supply to the cell. Mass flow in general is limited to a certain range and therefore the stoichiometry λ (λ = ratio of supplied reactant to chemically needed reactant) is also limited in dependency of the current density. At a low load point using maximum gas flow a higher stoichiometry can be realized than at high load points. Also the fuel cell itself will work properly with low gas pressure at low loads, but not at high loads.

Therefore it is on the one hand important to minimize the experimental work with consideration of these constraints and on the other hand to achieve the best possible parameter estimation. The aim of this article is to show and demonstrate a systematic method for investigation of a fuel cell, which performs both tasks simultaneously. It is known that the quality of the parameter estimation depends on the used measurement points and hence on the design of experiments (DoE). Therefore the issue can also be understood as to find the data set, which delivers the largest information content if experimental data are available or if not to provide an optimized experimental design for a PEMFC stack.

Box and Lucas [1] show in their work a mathematical theory for increasing the quality of least squares based parameter estimation by minimizing the covariance matrix of its solution. Several criteria are published in the literature [2] to evaluate this optimization. In this paper these criteria are compared and the most promising one is applied on a set of 405 test points, to find the set of 50 points which is most suitable for the parameter estimation of an empirical fuel cell voltage model. Moreover, it is investigated whether this

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Table 1
Typical factor levels of selected inputs in PEMFC characterization

Input Θ	Symbol	Unit	Factor levels
Current density	i	A/cm^2	10–20
Cathode gas pressure	p_c	bar	3–6
Anode gas pressure	p_a	bar	3–6
Relative humidity cathode gas	rh_c	–	3–6
Relative humidity anode gas	rh_a	–	3–6

optimized data set increases the quality of parameter estimation and therefore the stack model accuracy.

2. Design of experiments

2.1. Design methods

If the operating area is unrestricted and the number of fuel cell inputs are low, a full factorial DoE can be performed. Interactions as well as nonlinearities can be detected if more than three factor levels of each input are applied. But with a high number of inputs or factor levels the experimental effort rises exponentially. In Table 1 the typical number of factor levels is given. A full factorial design of experiments with the maximum values of each input would thus contain 25.920 experimental points.

If a system behaves linear within an unrestricted operation area a fractional factorial DoE can be chosen. That technique reduces the number of experiments compared to full factorial designs but is not able to detect interactions between inputs or nonlinear behaviour. Only a fraction of the edges of the hypercube, defined by the dimension of the operating area, is tested.

A trade-off between effort and ability to detect interactions and nonlinear behaviour gives a so-called central composed design. All edges of the hypercube are used. Additional, centre points and star points are added. These three designs are easy to construct, but all require an unrestricted operating area and are therefore not suited for global fuel cell characterization.

A first approach to design experiments in restricted areas is the so-called Box–Benken design. In this design the number of experiments is fixed and cannot be varied. The experiments are not placed in the edges of the operating area. More details about the mentioned and additional designs can be found in DoE literature, like Refs. [3–5]. More suited to the requirements for PEMFC characterization are optimal DoEs.

2.2. Optimal DoE

Since powerful computing has become affordable, more complex and powerful design criteria have been developed. The group of optimal criteria is able to design nonlinear DoEs with a low number of experiments in arbitrary restricted operating areas without being limited to a fixed number of experiments or factor levels.

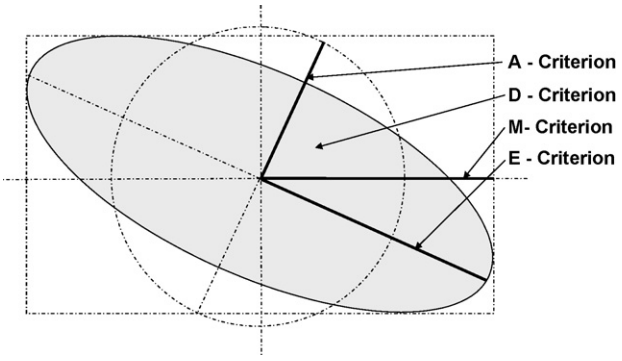

Fig. 1. Graphic interpretation of the optimal DoE criteria for minimizing the covariance matrix.

Table 2
Experimental conditions

State	Symbol	Operating range	
		Cathode	Anode
Pressure	p_c, p_a	1.05–2.5 bar	1.05–2.5 bar
Relative humidity	rh_c, rh_a	0.32–1	0.32–1
Temperature	T	75 °C	
Current density	i	0–2 A/cm^2	

Ref. [1] shows that the covariance matrix COV of the estimated model parameters ξ obtained by a least squares estimation can be approximated by the following equation:

$$\mathrm{COV} = (\mathrm{FIM}' \cdot \mathrm{FIM})^{-1}\sigma^2. \quad (1)$$

with the, in general unknown, variance σ^2 and FIM the Fisher information matrix. The FIM is a Φ-by-ϕ matrix of the partial derivative fim (Eq. (2)) of the θth model parameter ξ_θ for the dth set of experimental conditions Θ_d (Eq. (3)). Here d is the run index of the experimental conditions. It starts with 1 and ends at Φ. Additionally, θ is the index of the model parameters in the range from 1 to ϕ.

$$\mathrm{FIM} = \{\mathrm{fim}_{\theta d}\}, \quad \theta = 1 \ldots \phi, \quad d = 1 \ldots \Phi. \quad (2)$$

$$\mathrm{fim}_{\theta d} = \left[\frac{\partial\, \mathrm{fim}(\Theta, \xi)}{\partial \xi_\theta}\right]_{\theta_d}. \quad (3)$$

The basic idea of all optimal DoEs is the minimization of a quality criterion J related to COV or $(\mathrm{FIM}'\cdot\mathrm{FIM})^{-1}$ respectively. This optimization can be achieved by several design criteria [5].

The trace-criterion (A-criterion) (Eq. (4)) minimizes the trace,

$$J_A = \mathrm{trace}\left[\left(\mathrm{FIM}^{\mathrm{T}} \cdot \mathrm{FIM}\right)^{-1}\right] \quad (4)$$

the determinant-criterion (D-criterion) (Eq. (5)) minimizes the determinant,

$$J_D = \det\left[\left(\mathrm{FIM}^{\mathrm{T}} \cdot \mathrm{FIM}\right)^{-1}\right] \quad (5)$$

the eigenvalue-criterion (E-criterion) (Eq. (6)) minimizes the maximum eigenvalue (EIG),

$$J_E = \max\left\{\mathrm{EIG}\left[\left(\mathrm{FIM}^{\mathrm{T}} \cdot \mathrm{FIM}\right)^{-1}\right]\right\} \quad (6)$$

Table 3
Experimental details of campaigns 1–3 ($rh_{a/c}$ = 0.92)

Campaign					
1		2		3	
Gas pressure					
p_c (bar)	p_a (bar)	p_c (bar)	p_a (bar)	p_c (bar)	p_a (bar)
1.05	1.05	1.05	1.50	1.50	1.05
1.10	1.10	1.10	1.50	1.50	1.10
1.25	1.25	1.25	1.50	1.50	1.25
1.50	1.50	1.50	1.50	1.50	1.50
1.75	1.75	1.75	1.50	1.50	1.75
2.00	2.00	2.00	1.50	1.50	2.00
2.50	2.50	2.50	1.50	1.50	2.50