IEEE Access
Multidisciplinary | Rapid Review | Open Access Journal

Received 23 September 2022, accepted 14 October 2022, date of publication 21 October 2022, date of current version 31 October 2022.

Digital Object Identifier 10.1109/ACCESS.2022.3216364

METHODS

Parameter Individual Optimal Experimental Design and Calibration of Parametric Models

NICOLAI PALM[1], FLORIAN STROEBL[2], AND HERBERT PALM[2]
[1]Munich Center for Machine Learning (MCML), Ludwig-Maximilians University, 80799 Munich, Germany
[2]Institute for Sustainable Energy Systems (ISES), Munich University of Applied Sciences, 80335 Munich, Germany

Corresponding author: Florian Stroebl (florian.stroebl@hm.edu)

This work was supported in part by the German Federal Ministry for Economic Affairs and Climate Action under Grant 03EI4023A.

ABSTRACT Parametric models allow to reflect system behavior in general and characterize individual system instances by specific parameter values. For a variety of scientific disciplines, model calibration by parameter quantification is therefore of central importance. As the time and cost of calibration experiments increases, the question of how to determine parameter values of required quality with a minimum number of experiments comes to the fore. In this paper, a methodology is introduced allowing to quantify and optimize achievable parameter extraction quality based on an experimental plan including a process and methods how to adapt the experimental plan for improved estimation of individually selectable parameters. The resulting parameter-individual optimal design of experiments (pi-OED) enables experimenters to extract a maximum of parameter-specific information from a given number of experiments. We demonstrate how to minimize variance or covariances of individually selectable parameter estimators by model-based calculation of the experimental designs. Using the Fisher Information Matrix in combination with the Cramer-Raó inequality, the pi-OED plan is reduced to a global optimization problem. The pi-OED workflow is demonstrated using computer experiments to calibrate a model describing calendrical aging of lithium-ion battery cells. Applying bootstrapping methods allows to also quantify parameter estimation distributions for further benchmarking. Comparing pi-OED based computer experimental results with those based on state-of-the-art designs of experiments, reveals its efficiency improvement. All computer experimental results are gained in Python and may be reproduced using a provided Jupyter Notebook along with the source code. Both are available under https://github.com/nicolaipalm/oed.

INDEX TERMS Parametric models, parameter estimation, design of experiments, optimal experimental design, battery aging, computer experiment.

I. DESIGN OF COST INTENSIVE EXPERIMENTS

Models reduce systems of interest to their selected essential aspects. Parametric models allow to describe the behavior of cyber-physical systems by equations representing general relationships between input (independent or design) and output (dependent or target) variables, while the included parameters represent system- or material-specific individual properties. The identification of parameter values with high quality, therefore, plays a crucial role in many scientific disciplines such as material research [1], [2], pharmaceutics [3], [4] or mechatronics [5], [6].

The associate editor coordinating the review of this manuscript and approving it for publication was Bijoy Chand Chand Chatterjee.

A comprehensive methodological approach, underpinned by statistical tools, to maximize the amount of information in models to be drawn from a limited number of experiments was established a good 100 years ago by Ronald Fisher [7], [8]. The design of experiments (DoE) technique, based on Fisher's principles, still dominates the design of experimental plans today. A large body of work inspired by Fisher's basic approaches has extended the DoE approach to applications in areas such as parameter extraction of high-dimensional nonlinear models [9], [10], [11]. Often, however, this work has been primarily of theoretical interest as computational resources required to implement it have long been inadequate [12]. Conversely, this form of constraint has also prevented new approaches to optimized experimental designs

that focus on individual experimenter's aspects or goals such as the high-quality estimation of an individually selectable parameter within a parameterized model.

In this paper a new methodology is proposed a) providing an experimenter with information about achievable parameter extraction quality based on an experimental plan prior starting the experiments plus b) guidelines and tools how to adapt the experimental plan for improved estimation of individually selectable parameters. Based on this approach, an Optimal Experimental Design (OED) may be computed in order to maximize parameter specific information content of a parametric model as drawn from a given number of experiments. We call this new methodology *parameter individual Optimal Experimental Design* (pi-OED).

The paper is structured as follows: Following the introduction, in section II, the pi-OED methodology is sketched and explained in detail. II-A provides the statistical background of optimal experimental design in general including the statistical basis for parameter individual minimum variance estimation, followed by a step-by-step introduction of the generic workflow in II-B. To emphasize the potential of the pi-OED workflow, an example application in battery aging modelling is given in section III including computer experimental results that are compared to state-of-the-art experimental designs. The outcomes of this example are further discussed in section IV, before a conclusion is drawn and an outlook is presented in the final section V.

II. PI-OED METHODOLOGY

In this subsection, the methodology of pi-OED is presented including its statistical background, algorithm based workflow plus a brief sketch of required implementation tools.

pi-OED provides a framework to estimate parameter values within parametric models based on a twofold optimal behavior: The first aspect of optimality is related to a lower bound for variances and covariances of arbitrary parameter estimators that may be calculated for any given experimental design. Parameter estimators may be called (optimally) efficient if their variances and covariances approach this lower bound. The second aspect of optimality is related to an (optimal) experimental design minimizing the aforementioned lower bound. Some of the proofs may be found in the Appendix.

A. STATISTICAL BACKGROUND

Recall a statistical model $(\Omega, \mathcal{F}, P_\theta : \theta \in \Theta)$ to consist of a set Ω, a σ-algebra $\mathcal{F}$ on Ω, a (parameter-)set Θ and for each $\theta \in \Theta$ a probability measure P_θ on Ω. Equipping Ω with the structure of a measurable space, an estimator for θ is then a measurable function $T : \Omega \to \Theta$.

For the rest of this subsection, let

$$(\mathbb{R}^n, \mathcal{B}^n, P_\theta : \theta \in \Theta) \tag{1}$$

be a statistical model with

1) $\Theta \subset \mathbb{R}^m$ open
2) such that each P_θ has a continuous density function $\rho_\theta : \mathbb{R}^n \to \mathbb{R}$ and
3) the likelihood function

$$\rho : \Theta \times \mathbb{R}^n \to \mathbb{R}, (\theta, x) \mapsto \rho_\theta(x) \tag{2}$$

being strictly positive and
4) admitting continuous partial derivatives $\frac{\partial}{\partial\theta_i}\rho$ and second partial derivatives $\frac{\partial^2}{\partial\theta_i\partial\theta_j}\rho$ for all $i, j = 1, \ldots, m$.

Notation:

1) Given a probability measure P_θ on some space X and a real valued measurable function T on X, we denote by $E_\theta(T) = \int_X T dP_\theta$ the expected value of T.
2) Similar to 1), we denote by

$$\mathrm{var}_\theta(T) = E_\theta((T - E_\theta(T))^2) \tag{3}$$

the variance of T.
3) More general, if $T = (T_i)_{i=1,\ldots,n}$ takes values in $\mathbb{R}^n$, we denote by $E_\theta(T) = (E_\theta(T_i))_{i=1,\ldots,n}$ its componentwise expected value and by

$$C_\theta(T) = (\mathrm{cov}_\theta(T_i, T_j))_{i,j=1,\ldots,n} \tag{4}$$

its covariance matrix with

$$\mathrm{cov}_\theta(T_i, T_j) = E_\theta[(T_i - E_\theta(T_i))(T_j - E_\theta(T_j))]. \tag{5}$$

4) For the remainder of the text, we will assume every subset $U \subset \mathbb{R}^n$ to be endowed with the standard Borel σ-algebra structure $\mathcal{B}^n$.

Example 1 (Motivating Example): We consider an experimental setup consisting of a design space X of experimental design options. Conducting a single experimental design $x \in X$ leads to a d-dimensional (result) vector $y \in \mathbb{R}^d$ of real numbers. We refer to an experiment $\xi = (x_1, \ldots, x_k) \in X^k$ as a vector of k experimental designs.

Assume we conduct an experiment consisting of n experimental designs $x_1, \ldots, x_n \in X$. Assume the result being not deterministic but multivariate normal distributed with mean zero and covariance matrix $\sigma^2\mathbb{1}_d$ indicating the experimental error. In order to predict the result y at some new design x we choose a parametric model, i.e., a set Θ and, for each $\theta \in \Theta$ a function $f_\theta : X \to \mathbb{R}^d$, where we assume some $f_{\theta_{true}}$ to be the *true* model with $\theta_{true} \in \Theta$. We then strive to calculate θ_{true} under the use of the evaluations $y_1, \ldots, y_n \in \mathbb{R}^d$. In mathematical terms, we consider the statistical model

$$(\mathbb{R}^{dn}, \mathcal{B}^{dn}, \mathcal{N}((f_\theta(x_i))_{i=1,\ldots,n}, \sigma^2\mathbb{1}_{dn}) : \theta \in \Theta) \tag{6}$$

with $\mathcal{N}((f_\theta(x_i))_{i=1,\ldots,n}, \sigma^2\mathbb{1}_{dn})$ being a multivariate normal distribution with mean $(f_\theta(x_i))_{i=1,\ldots,n}$ and covariance matrix $\sigma^2\mathbb{1}_{dn}$. For this statistical model, we are seeking an estimator assigning the resulting vector of n experimental designs to an estimate for the parameter.

1) PARAMETER ESTIMATION

Recall an estimator $T : \mathbb{R}^n \to \Theta \subset \mathbb{R}^m$ to be unbiased if

$$E_\theta(T) = (E_\theta(T_i))_{i=1,\ldots,n} = \theta \tag{7}$$

for all $\theta \in \Theta$. In words, the average of estimates will yield the true parameter.

Among the class of unbiased estimators we are interested in the estimators with (componentwise) minimum variance, i.e. $\mathrm{var}_\theta(T_i)$ is minimal among all unbiased estimators for all $i = 1, \ldots, n$.

In general, a minimum variance unbiased estimator may not be found or even exist. However, there is a convenient estimator which exists under mild conditions and satisfies highly desirable properties (proposition 9 and theorem 10).

Definition 2 (Maximum Likelihood Estimator): An estimator $T : \mathbb{R}^n \to \Theta \subset \mathbb{R}^m$ is a maximum likelihood estimator (MLE) if T maximizes the likelihood function ρ, i.e. if

$$\rho(T(x), x) = \max_{\theta \in \Theta}(\rho(\theta, x)) \tag{8}$$

for all $x \in \mathbb{R}^n$.

It is well known that the maximum likelihood estimator agrees with the least square error estimator in case of a multivariate normal distribution (see for instance 8.9 in [13] or [14], [15]).

Example 3 (Continuation of Example 1): In example 1 maximizing the likelihood function is equivalent to minimizing the mean square error function

$$\Theta \to \mathbb{R}, \theta \mapsto \sum_{i=1}^{n} \sum_{j=1}^{d} (y_{i,j} - f_\theta(x_i)_j)^2, \tag{9}$$

with $y_{i,j}$ and $f_\theta(x_i)_j$ defined as j-th component of evaluation y_i and $f_\theta(x_i)$, respectively.

2) FISHER INFORMATION MATRIX

Under convenient assumptions, there exists a general lower bound for the covariance matrix of each unbiased estimator (theorem 6), which approximately is attained by the maximum likelihood estimator (theorem 10). This lower bound is given by the inverse of the *Fisher Information Matrix*. For the rest of this chapter, we assume $\ln(\rho_\theta)$ to exist and to admit first and second partial derivatives.

Definition 4 (Fisher Information Matrix): The Fisher Information Matrix (FIM) $I(\theta) \in M_n(\mathbb{R})$ is componentwise defined by

$$I(\theta)_{ij} = -E_\theta\left(\frac{\partial^2 \ln \rho_\theta}{\partial\theta_i \partial\theta_j}\right). \tag{10}$$

Its inverse (if existing) is called the *Cramer-Rao lower bound (CRLB).*

Example 5 (Continuation of Example 1): In example 1 the FIM is given by

$$I(\theta)_{ij} = \frac{1}{\sigma^2} * \sum_{l=1}^{m} \left(\frac{\partial}{\partial\theta_i} f_\theta(x_l) * \frac{\partial}{\partial\theta_j} f_\theta(x_l)\right). \tag{11}$$

Observe that the FIM depends on the experimental designs.

Proof: Example 3.9 in [13] (See formula (3.33)) □

3) CRAMER RAO LOWER BOUND

In the following, we assume the FIM corresponding to the statistical model to be invertible for all parameters and that the MLE exists whenever it is used.

Theorem 6: Assume the likelihood functions to satisfy the regularity condition

$$E_\theta\left(\frac{\partial \ln \rho_\theta}{\partial\theta_i}\right) = 0 \tag{12}$$

for all i and $\theta \in \Theta$. Let $T : \mathbb{R}^n \to \Theta$ be an unbiased estimator. Then, the covariance matrix of the estimator $C_\theta(T)$ with respect to θ satisfies

$$C_\theta(T) - I^{-1}(\theta) \tag{13}$$

is positive semi-definite for every $\theta \in \Theta$.

Furthermore, an unbiased estimator may be found that attains the bound in that $C_\theta(T) = I^{-1}(\theta)$ if and only if

$$\frac{\partial \ln \rho_\theta}{\partial\theta}(x) = I(\theta)(g(x) - \theta) \tag{14}$$

for some function $g : \mathbb{R}^n \to \mathbb{R}^m$. That estimator, which is the minimum-variance unbiased estimator, is $T = g$.

Such an estimator is called efficient.

Proof: Theorem 3.2 in [13] □

The lower bound derived in the previous theorem yields a lower bound for the variance of each individual parameter estimator.

Corollary 7: Suppose we are in the situation of theorem 6. Then, the variance of the parameter estimation of the i-th component is greater or equal than the i-diagonal entry of the inverse of the FIM, i.e.

$$\mathrm{var}_\theta(T_i) \geq I^{-1}(\theta)_{ii} \tag{15}$$

for all $\theta \in \Theta$.

Proof: See Appendix! □

Example 8 (Continuation of Example 1): The statistical model of example 1 satisfies the regularity condition of theorem 6.

If an efficient estimator exists, it may be difficult to determine it. However, in that case we can always use the maximum likelihood estimator:

Proposition 9: If an efficient estimator exists, then it is provided by the maximum-likelihood estimator.

Proof: See Appendix! □

4) ASYMPTOTIC EFFICIENCY OF THE MLE

The maximum likelihood estimator and the FIM are closely related:

Theorem 10 (Aysmptotic Efficiency of the MLE): Assume the maximum likelihood estimator exists and is measurable. Assume further the likelihood functions satisfy the regularity condition

$$E_\theta\left(\frac{\partial \ln(\rho_\theta)}{\partial\theta_i}\right) = 0 \tag{16}$$

for all $\theta \in \Theta$. Then, the maximum-likelihood estimator T is asymptotically (multivariate-)normally distributed according to

$$T \overset{a}{\sim} \mathcal{N}(\theta_{true}, I^{-1}(\theta_{true})), \tag{17}$$

where θ_{true} denotes the true value of the underlying probability measure on $\mathbb{R}^n$. In particular, the maximum likelihood estimator is asymptotically efficient and, in particular, asymptotically unbiased.

Proof: Theorem 7.1 in [13] □

5) PROPERTIES OF THE FISHER INFORMATION MATRIX

In this section we state some properties of the FIM. They complement the theory above in terms of practical applications with some of the proofs to be found in the Appendix.

Proposition 11 (FI Positive Semi-Definite): The FIM $I(\theta)$ is positive semi-definite for every $\theta \in \Theta$.

Proof: Problem 3.10 in [13] □

We are mainly interested in the properties of the FIM when adding experimental designs. Let us assume in example 1 that we conduct another experiment with e experimental designs $x_{n+1}, \ldots, x_{n+e}$. We wonder if joining both experiments may lead to a higher variance in the individual parameter estimations in comparison with the initial experiment $(x_1, \ldots, x_n)$. Intuitively, this should not be possible. Considering the CRLB as the covariance matrix of the MLE (in the sense of theorem 10), the below proposition guarantees that adding experimental designs will decrease the variance of the individual parameter estimations. In particular, minimizing a specific entry on the diagonal of the CRLB by adding experimental designs won't increase its other diagonal entries.

Proposition 12: Let

$$(\mathbb{R}^{n'}, \mathcal{B}^{n'}, P'_\theta : \theta \in \Theta) \tag{18}$$

be another statistical model satisfying the assumptions stated at the beginning of this chapter. We obtain the statistical model

$$(\mathbb{R}^n \times \mathbb{R}^{n'}, \mathcal{B}^{n+n'}, P_\theta \otimes P'_\theta : \theta \in \Theta) \tag{19}$$

with density function of $P_\theta \otimes P'_\theta$ given by the product of the density functions of P_θ and P'_θ. Denote by $\tilde{I}(\theta)$ its corresponding FIM at some $\theta \in \Theta$.

Then, $\tilde{I}(\theta)$ is positive definite and

$$I^{-1}(\theta) - \tilde{I}^{-1}(\theta)$$

is positive semi-definite.

In particular,

$$I^{-1}(\theta)_{ii} \geq \tilde{I}^{-1}(\theta)_{ii}$$

for all i.

Proof: See Appendix! □

Theorems 6 and 10 highly depend on the invertibility of the FIM. Therefore, we investigate the invertibility in the special case of our motivating example.

Proposition 13 (Invertibility of the FI for White Gaussian Noise Models): Assume $d = 1$ in example 1 and the f_θ are continuously differentiable. Then, the FIM is invertible at θ if and only if without loss of generality the continuous differentiable map

$$F : \Theta \to \mathbb{R}^m, \theta \mapsto (f_\theta(x_i)_{i=1,\ldots,m})$$

is a local diffeomorphism in an open neighbourhood around θ.

Proof: See Appendix! □

Roughly speaking, the FIM is invertible if and only if in case of data noise absence the determination of model parameters from the given experiment is (locally) unique.

Last, we investigate the existence of the maximum likelihood estimation. We can prove the existence of the MLE under some convenient assumptions.

Proposition 14: Assume $\Theta \subset \mathbb{R}^m$ is bounded and the likelihood function ρ admits a continuous extension $\bar{\rho}$ to the closure $\bar{\Theta}$ of Θ, i.e. the diagram

$$\begin{array}{ccc} \Theta \times X & \subset & \bar{\Theta} \times X \\ \downarrow{\scriptstyle \rho} & \swarrow{\scriptstyle \bar{\rho}} & \\ \mathbb{R} & & \end{array}$$

commutes. Then, there exists a maximum of

$$\bar{\rho}(-, x) : \bar{\Theta} \to \mathbb{R}, \theta \mapsto \bar{\rho}(\theta, x)$$

for all $x \in X$. In particular, assuming the maximum of $\bar{\rho}(-, x)$ to be in Θ, then, $\rho(-, x)$ attains its maximum and the MLE exists.

Proof: See Appendix! □

In practice, most of the above assumptions are often satisfied or may be assumed to be satisfied.

6) SUMMARY

Experimental errors may cause parameter estimation errors of the assigned model. A useful class of estimators contains those elements whose expected value yields the true parameter value (i.e. unbiased estimators). This class of unbiased estimators admits a general lower bound for their covariance matrices, the CRLB (theorem 6). The covariance matrix of some estimator is a suitable measure of how much its parameter estimation suffers from the error in the experimental results. Thus, we strive to find an unbiased estimator with minimal variance. Because the maximum likelihood estimator (MLE) is

- efficient if an efficient estimator exists at all (proposition 9)
- approximately efficient (theorem 10)

it is our estimator of choice when we cannot find the unbiased minimum variance estimator by hand.

In general, the FIM depends on the choice of experimental designs. Accordingly, a suitable choice of experimental designs allows us to (asymptotically) quantify and pro-actively minimize the covariance of the maximum likelihood estimation.

B. GENERIC WORKFLOW

In this subsection, the pi-OED workflow as shown in Figure 1 is introduced step-by-step. The iterative generic process faces the challenge of finding model parameters and improving model quality in an effective and efficient way. Building on

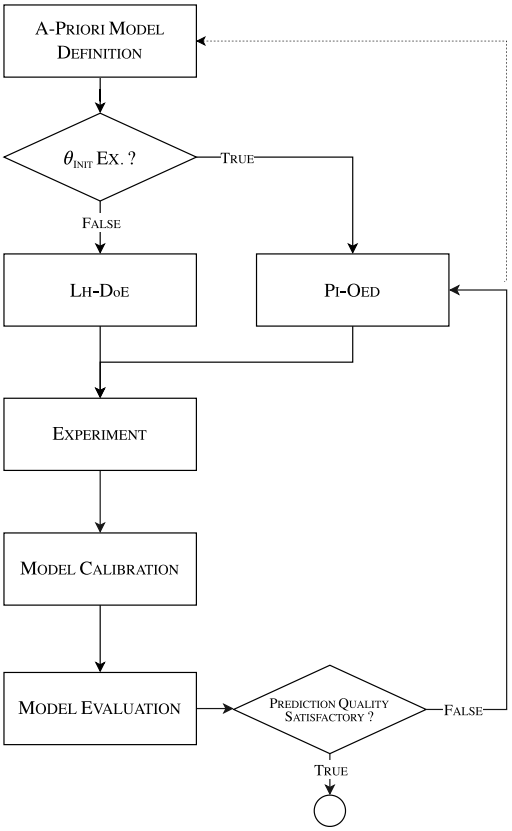


FIGURE 1. Generic pi-OED workflow.

the theoretical foundations presented in II-A, it can be applied to any kind of statistical models satisfying a few convenient assumptions.

1) A PRIORI MODEL DEFINITION

Aim of the 'A priori model definition' block is to translate the data of an experimental setup into the mathematical language, i.e. into statistical models.

We consider an experimental setup consisting of a design space X of experimental design options. We assume the design space $X \subset \mathbb{R}^m$ to consist of real valued m-dimensional vectors and to be a bounded cross product of intervals, i.e. each coordinate is element of a bounded interval. Conducting a single experimental design $x \in X$ leads to a n-dimensional (result) vector $y \in \mathbb{R}^n$ of real numbers. In addition, the output y of a measurement is randomly distributed. We refer to an experiment $\xi = (x_1, \ldots, x_k) \in X^k$ as a vector of k experimental designs.

For each experiment $\xi = (x_1, \ldots, x_k)$ we make an assumption about the underlying distribution of its joint experimental output $(y_1, \ldots, y_k)$ in form of a statistical model

$$((\mathbb{R}^n)^k, \mathcal{B}^{nk}, P_{\theta,\xi} : \theta \in \Theta). \tag{20}$$

This can be done by either testing the underlying distribution using statistical tests such as a Shapiro-Wilk-Test [16], by using prior knowledge, or simply by assumption. We further demand each statistical model to satisfy the assumptions of theorem 6. Furthermore, we assume the MLE to exist whenever it is used. In practice, a suitable criterion is given by proposition 14.

Observe that the parameter space Θ is supposed to not depend on the experiment ξ, i.e. for every experiment ξ, Θ remains the same. Accordingly, we strive to estimate the value of a *true* (independently from any experiment existing) parameter $\theta_{true} \in \Theta$.

2) DOES AN INITIAL PARAMETER ESTIMATE EXIST?

Given the statistical models for all experiments derived in the a priori model definition block, an initial θ_{init} guess may exist based on previously conducted experiments or additional knowledge about the underlying model. If so, we continue with the pi-OED DoE block. If not, we continue with LH-DoE.

3) LH-DoE

Assume an initial guess for the parameter does not exist. Then, Latin Hypercube (LH) based Design of Experiments (DoE) [17] defines a suitable set of experimental designs allowing us to effectively and efficiently achieve a base for an initial parameter guess. This sampling-like approach results in an experiment $\xi_{LH} = (x_1, \ldots, x_k)$ that represents the variability of design space by stratification and experimental design selection according to a predetermined number k of available individual experimental designs. The LH-DoE calculation is possible because the design space $X \subset \mathbb{R}^m$ is a bounded cross product of intervals.

4) EXPERIMENT

Having defined the experiment $\xi = (x_1, \ldots, x_k)$ by LH-DoE or pi-OED-DoE, we now run it to obtain a corresponding vector of experimental outputs $(y_1, \ldots, y_k) \in (\mathbb{R}^n)^k$.

5) MODEL CALIBRATION

Model calibration assigns specific parameter values θ to the underlying statistical models. Joining all experiments and experimental outputs of previously conducted experiments, we obtain an experiment $\xi = (x_1, \ldots, x_l)$ of l individual experimental designs with corresponding experimental output $(y_1, \ldots, y_l)$. We calculate the maximum likelihood estimation of $(y_1, \ldots, y_l)$ (see 2) with respect to the statistical model

$$((\mathbb{R}^n)^l, \mathcal{B}^{nl}, P_{\theta,\xi} : \theta \in \Theta), \tag{21}$$

i.e. we solve the maximization problem

$$\underset{\theta \in \Theta}{\operatorname{argmax}}(\rho_\xi(\theta, (y_1, \ldots, y_l))), \tag{22}$$

where

$$\rho_\xi : \Theta \times (\mathbb{R}^n)^l \to \mathbb{R}$$

denotes the likelihood function corresponding to the statistical model. This problem can be solved either by numerical

calculation or, in some cases, in closed form, leading to a parameter estimation of θ.

6) MODEL EVALUATION

Model Evaluation deals with two basic questions: (i) Are models useful within a defined measure of forecast quality and (ii) are determined parameter values precisely enough estimated. Given the parameter estimation of θ, we seek to evaluate the prediction capability (quality of forecast) of the corresponding probability measures, i.e. determine whether we have chosen sufficiently useful statistical models. Evaluation criteria and their according indicator base are widely known and problem specific [18], [19], [20], [21].

Randomness in measurement results leads to a randomness in the parameter estimation, i.e. conducting a single experiment multiple times leads to different parameter estimations. Accordingly, we need to evaluate the quality of the parameter estimation, i.e. quantify the uncertainty of our parameter estimation.

The inverse of the FIM at θ is a suitable approximate of the covariance matrix of the parameter estimator (theorem 6). In return, the covariance matrix is a suitable measure of the randomness of the parameter estimate. Therefore, we first check the invertibility of the FIM $I(\theta, \xi)$ corresponding to the statistical model

$$((\mathbb{R}^n)^l, \mathcal{B}^{nl}, P_{\theta,\xi} : \theta \in \Theta) \tag{23}$$

by calculating its determinant. If the determinant is non-zero, we derive an applicable criterion by considering the relative expected standard deviations

$$\sigma_{rel,i} = \frac{\sqrt{I^{-1}(\theta, \xi)_{ii}}}{\theta_i} \tag{24}$$

for $i = 1, \ldots, n$. The resulting values are appropriate uncertainty measures for individual components of the estimated parameter θ.

7) DECISION ON MODEL QUALITY

Based on the calculated metrics in the model evaluation block we face three options:

1) Reject the underlying statistical models. Then, we have to re-define the underlying statistical models in the a priori model definition block.
2) Accept the underlying statistical models but reject the parameter estimation quality. Then, we continue with pi-OED for improving selected parameter quality.
3) Accept the model P_θ. Then, we end the workflow.

In general, accurate parameter estimation quality is required before underlying statistical models can be rejected. A non-invertible FIM may reflect the lack of experimental information (see proposition 13), and the experimenter is advised to discard the quality of the parameter estimate. The amount or quality of data may be considered insufficient. In the case of an invertible FIM, we usually accept relative expected standard deviations below a predefined limit (confidence level), e.g. five percent.

8) pi-OED DoE

pi-OED DoE allows to (i) individually select those parameters not yet meeting the desired level of quality and (ii) calculate at a predefined number of available experimental designs the experiment maximizing the information content with respect to estimation of the selected parameters.

We are given an initial parameter guess θ, the merged experiment $\xi = (x_1, \ldots, x_l)$ and corresponding experimental outputs $(y_1, \ldots, y_l)$ of previously conducted experiments.

Recall that the inverse of the FIM at the parameter θ is an approximate of the covariance matrix of the maximum-likelihood estimation in the sense of theorem 10 and a lower bound to every unbiased parameter estimation (Theorem 6). Accordingly, its diagonal entries are approximates of the variances of the individual components of the parameter. In order to minimize the variance of a parameter component, we design the parameter individual optimal experiment (pi-OED).

We choose an index i whose corresponding parameter component's θ_i uncertainty (i.e. parameter estimator variance) we wish to minimize. Given a number of additional experimental designs s, the pi-OED experiment $\xi_{pi} = (x_{l+1}, \ldots, x_{l+s})$ is given by solving the minimization problem

$$\xi_{pi} = \underset{x_{l+1},\ldots,x_{l+s}}{\operatorname{argmin}} \; I^{-1}(\theta, (x_1, \ldots, x_l, x_{l+1}, \ldots, x_{l+s}))_{ii}. \tag{25}$$

Note that according to theorem 12, the other entries on the diagonal (i.e. the expected variance of the respective parameter component estimation) also decrease as more experimental designs are added. Resulting in an parameter-individual optimal experimental design, we then proceed with step II-B4 and continue in our workflow.

III. EXAMPLE pi-OED APPLICATION IN PARAMETRIC MODELS WITH WHITE GAUSSIAN NOISE

In the following section, we apply the presented pi-OED workflow to a cost-intensive technical problem. For demonstration purposes, we have chosen the characterization of the aging behavior of lithium-ion battery cells as an example. Identifying an adequate model and quantifying associated (for the example fixed but for its experimenter a priori unknown) model parameters with sufficient confidence level is essential for tasks such as battery development or system design optimization of battery storage systems [22], [23].

Chapter III is structured as follows: III-A describes the fundamental workflow for battery aging characterization following in step-by-step mode the generic pi-OED workflow as shown in Figure 1 and indicating computer experimental results for the chosen example. Repeatedly simulating individual experiments along this workflow allows to calculate statistical distributions according to a bootstrapping approach [24], specifically with respect to mean values and variances. Results will be compared to their theoretically expected values according section II. Within III-B we then benchmark pi-OED versus state-of-the-art DoE

results. Source code and computer experimental results can be found in https://github.com/nicolaipalm/oed/blob/master/notebooks/pi_workflow_example.ipynb.

A. WORKFLOW WITH RESULT AND STATISTICS

Capacity fade due to battery storage and operation, is often used as an indicator for the degradation of lithium-ion batteries. The usual approach of considering calendrical and cyclical aging as linearly independent influences on the capacity loss leads to the following general model structure [25], [26], [27], [28]:

$$Q_{loss} = Q_{loss}^{cal} + Q_{loss}^{cyc}, \tag{26}$$

where Q_{loss} defines the total capacity loss of the battery compared to its initial value, while Q_{loss}^{cal} and Q_{loss}^{cyc} denote the aging contribution of calendrical and cyclic aging, respectively. This model allows to divide the characterization of aging into two independent experimental studies. For our example, we focus exclusively on the calendrical aging part of the model with Q_{loss}^{cal} as proposed by Muehlbauer et al. [28]:

$$Q_{loss}^{cal}(T, SoC, t) = x_{ref} \cdot d_T^{cal}(T) \cdot d_{SoC}^{cal}(SoC) \cdot t^{z_{cal}}, \tag{27}$$

with

$$d_T^{cal} = e^{-\gamma_T^{cal} * \left(\frac{1}{T} - \frac{1}{T_{ref}}\right)}, \tag{28}$$

$$d_{SoC}^{cal} = \left(\frac{SoC}{SoC_{ref}}\right)^{\frac{1}{\gamma_{SoC}^{cal}}}, \tag{29}$$

and

$$x_{ref} = \left(\frac{1 - EOL_C}{(t_{end})^{z_{cal}}}\right), \tag{30}$$

where the ambient storage temperature T, the storage state of charge SoC and the storage time t represent the independent variables of the model. SoC_{ref} and T_{ref} define reference values of SoC and ambient temperature, respectively. EOL_C defines the End of life (EOL) capacity of a battery cell that in our example is defined by a value of $EOL_C = 90\%$, reached when a cell is stored at $SoC_{ref} = 50\%$ and $T_{ref} = 296.15K$ for $t_{end} = 520$ days. In our model, the material and aging mechanism specific parameters γ_{SoC}^{cal}, γ_T^{cal} (stress exponents) and z_{cal} (exponential factor for time) represent the parameter vector θ components:

$$\theta = (\theta_0, \theta_1, \theta_2) = (\gamma_{SoC}^{cal}, \gamma_T^{cal}, z_{cal}). \tag{31}$$

The parameter values θ_i with $i = 0, 1, 2$ shall be identified on the basis of experiments. Following the pi-OED workflow, we will simulate computer experimental results instead of performing real experiments. This enables us to quantify means and variances of experimental results (by bootstrapping) and comparing those results to theoretical predictions.

The parametric model according to equations (26)-(30) represents the family of models

$$f_\theta = Q_{loss}^{cal} \tag{32}$$

for the pi-OED workflow demonstration. Temperature (T) ranges from $T_l = 279.15K$ to $T_u = 333.15K$. State of charge (SoC) ranges from $SoC_l = 0.05$ to $SoC_u = 1$. Unless otherwise specified, time (t) and temperatures (T) are positive real number, measured in days or Kelvin, respectively.

An experimental design is characterized by temperature T and state of charge SoC. The design space is, therefore, given by the cross product of intervals

$$X = (T_l, T_u) \times (SoC_l, SoC_u).$$

For each experimental design we conduct measurements at $t_0 = 7, t_1 = 35, t_2 = 63, t_3 = 119, t_4 = 175$ and $t_5 = 231$ of days after initialization of the batteries. Accordingly, conducting an experimental design results in a vector $y \in \mathbb{R}^6$.

Based on prior knowledge the parameter space is estimated by

$$\Theta = (0.1, 10) \times (0, 10000) \times (0, 1).$$

In mathematical terms, we consider the parametric function

$$f_\theta : (T_l, T_u) \times (SoC_l, SoC_u) \times \mathbb{R}_+ \to [0, 1]$$

with $\theta \in \Theta$.

The measurement errors of conducting single experimental designs are assumed independent and identical normally distributed (iid) with zero mean and standard deviation $\sigma = 0.002$. Accordingly, conducting n experimental designs results in a multivariate normal distribution with covariance matrix $\sigma^2 \mathbb{1}_n$.

In our example we define the *true* (and for the experimenter a priori unknown) model parameter

$$\theta_{true} = (4, 2300, 0.8).$$

Conducting an experimental design $x = (T, SoC)$ yields the result vector

$$(f_{\theta_{true}}(T, SoC, t_0), \ldots, f_{\theta_{true}}(T, SoC, t_5)) \in \mathbb{R}^6$$

superimposed by a random error distributed according to $\mathcal{N}(0, \sigma^2 \mathbb{1}_6)$. The reader is encouraged to validate the results with different parameter θ_{true}.

In the following, optimization tasks are solved by the differential evolution global optimization algorithm [29], [30]. More precisely, we phrased optimization tasks as minimization problems and used the algorithmic implementation provided in the Python module *scipy.optimize.differential_evolution* of the SciPy 1.8.1 release with maxiter set to 1000 and tolerance set to 1e-5.

1) A PRIORI MODEL DEFINITION

According to the above experimental setup, given an experiment $\xi = (x_1, \ldots, x_k)$ with experimental designs

$$x_i = (T_i, SoC_i) \in X,$$

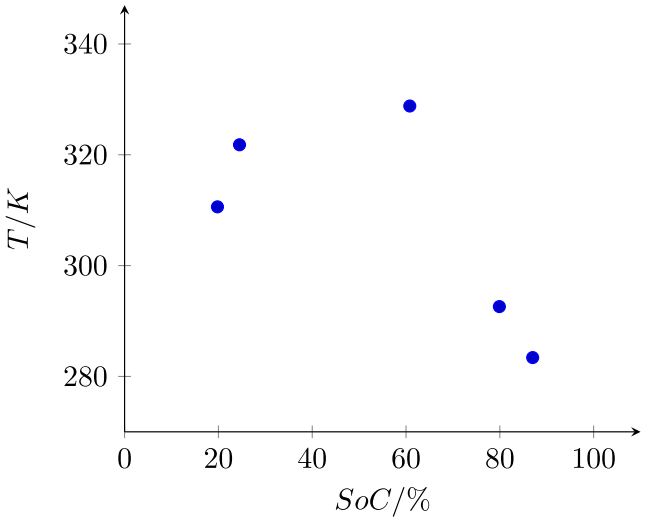


FIGURE 2. Latin Hypercube DoE.

we obtain the statistical model

$$(\mathbb{R}^{6k}, \mathcal{B}^{6k}, \mathcal{N}((f_\theta(T_i, SoC_i, t))_{i=1,\dots,k}, \sigma^2 \mathbb{1}_{6k}) : \theta \in \Theta)$$

with parameter space

$$\Theta = (0.1, 10) \times (0, 10000) \times (0, 1)$$

where we write

$$f_\theta(T_i, SoC_i, t) = (f_\theta(T_i, SoC_i, t_j))_{j=0,\dots,5}.$$

By example 8 this model satisfies the assumptions of theorem 6. Observe that all statistical models inherit the same parameter space Θ. Experimentally determining the (global) parameter θ calibrates the aging model f_θ.

2) INITIAL DESIGN OF EXPERIMENT

Initially, we assume no prior knowledge about the value of model parameter θ to exist and an ability to conduct an experiment consisting of $k = 5$ experimental designs. Therefore, the initial experiment is designed by the Latin Hypercube (LH) approach with five samples out of X resulting in the experiment

$$\xi_{LH} = ((T_1, SoC_1), \dots, (T_5, SoC_5)).$$

Figure 2 indicates the calculated LH experiment with each blue dot representing an individual experimental design.

3) CONDUCT EXPERIMENTS

We conduct the Latin Hypercube experiment ξ_{LH}. This results in 30 experimental data points $y_{i,j}$ based on measurements for $i = 1, \dots, 5$ experimental designs measured at $j = 0, \dots, 5$ points in time.

4) MODEL CALIBRATION

The statistical model joining all experiments is given by

$$(\mathbb{R}^{30}, \mathcal{B}^{30}, \mathcal{N}((f_\theta(T_i, SoC_i, t))_{i=1,\dots,5}, \sigma^2 \mathbb{1}_{30}) : \theta \in \Theta).$$

The closure of Θ is given by $[0.01, 10] \times [0, 10000] \times [0, 1]$. Using the formulas for f_θ, we observe that f_θ can be extended to the closure. In particular, the likelihood function corresponding to the statistical model admits a continuous extension to the closure. Thus, this extension attains its maxima in the sense of proposition 14. Assuming the maximum is not at the boundary $\{0.01, 1\} \times \{0, 10000\} \times \{0, 1\}$ we conclude the MLE to exist.

According to example 5, the maximum likelihood estimate for θ of the experimental output $(y_{i,j})_{i=1,\dots,5,j=0,\dots,5}$ is given by the least square error estimate

$$\operatorname*{argmin}_\theta \sum_{i=1}^{5} \sum_{j=0}^{5} (y_{i,t} - f_\theta(T_i, SoC_i, t_j))^2.$$

That parameter θ_{init} is calculated by applying the differential evolution optimizer described above to the objective function

$$\theta \mapsto \sum_{i=1}^{5} \sum_{j=0}^{5} (y_{i,t} - f_\theta(T_i, SoC_i, t_j))^2$$

where the search space is set to Θ. The resulting initial parameter estimate is then given by

$$\theta_{init} = (3.90, 2351.57, 0.81).$$

Note that this parameter estimate is non-deterministic. Bootstrapping with $N_{rep} = 1000$ fold resampling within our 30-dimensional sample space allows us to quantify the statistical distribution of the parameter estimator. Figures 3(a)-(c) show the normalized distributions of the estimated parameter vector component distributions after initial LH DoE as obtained via bootstrapping. The numbers are scaled evenly with the *true* parameter value as the center and a range of $\pm 40\%$ around this value. The probability density function (pdf) of normal distributions with mean values defined by the true parameter values θ_i and variances by respective diagonal entries of the CRLB is shown in red within the according sub-figures. Note that bootstrapping is not part of the regular workflow but serves as additional validation in this particular example.

5) MODEL EVALUATION

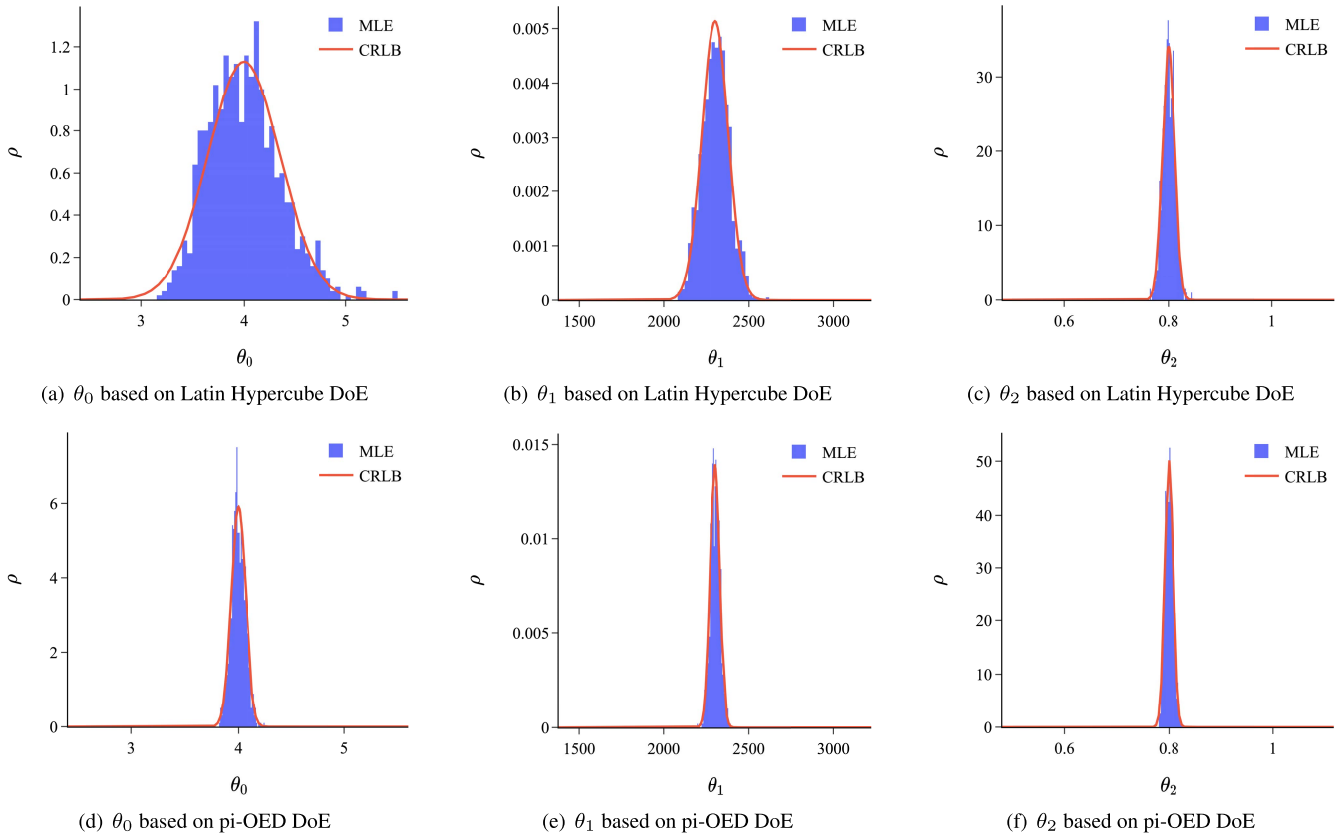
To evaluate the predictive ability of the now initially calibrated model, we choose the mean absolute error (MAE) as a performance measure determined by the leave-one-out cross-validation (LOOCV) method [31], [32]. This indicator is suitable for situations where only little data is available. In our example, the calculated MAE is 0.0024. Observe this error to be in the range of the known measurement error $\sigma = 0.002$.

The determinant of the FIM corresponding to

$$(\mathbb{R}^{30}, \mathcal{B}^{30}, \mathcal{N}((f_\theta(T_i, SoC_i, t))_{i=1,\dots,5}, \sigma^2 \mathbb{1}_{30}) : \theta \in \Theta)$$

is given by

$$\det(I(\theta_{init}, \xi_{LH})) = 180.23 > 0.$$



(a) θ_0 based on Latin Hypercube DoE (b) θ_1 based on Latin Hypercube DoE (c) θ_2 based on Latin Hypercube DoE

(d) θ_0 based on pi-OED DoE (e) θ_1 based on pi-OED DoE (f) θ_2 based on pi-OED DoE

FIGURE 3. Comparison of estimated parameter vector component distributions for the chosen example (a)-(c) after initial LH DoE and (d)-(f) after pi-OED. Blue bars indicate bootstrap based MLE histograms, red lines indicate CRLB based distributions.

TABLE 1. Relative expected standard deviation of θ_{init}.

$\theta_{init,i}$	θ_0	θ_1	θ_2
$\sigma_{rel,i}$	6.1%	2.0%	1.3%

In particular, the FIM $I(\theta_{init}, \xi_{LH})$ is invertible. According 6, the CRLB at the initially estimated parameter θ_{init} is given by

$$I^{-1}(\theta_{init}, \xi_{LH}) = \begin{pmatrix} 5.7\text{e}{-02} & -2.0\text{e}{+00} & -7.7\text{e}{-05} \\ -2.0\text{e}{+00} & 2.2\text{e}{+03} & 4.0\text{e}{-01} \\ -7.7\text{e}{-05} & 4.0\text{e}{-01} & 1.2\text{e}{-04} \end{pmatrix}$$

quantifying the relative expected standard deviation $\sigma_{rel,i}$ for each parameter component $\theta_{init,i}$ with respect to the initial parameter estimate θ_{init}. $\sigma_{rel,i}$ results are shown in Table 1.

6) PREDICTION QUALITY SATISFACTORY?

In our example, we request a relative expected standard deviation below five percent (i.e. confidence level). According Table 1, we accept estimation of θ_1 and θ_2 while refusing quality of θ_0.

However, even at this point we recognize the LOOCV error to be in the range of the measurement error. This reflects a suitable choice of the underlying statistical model (i.e. the aging model). This is not surprising since the underlying simulation of experiments is given by $f_{\theta_{true}}$.

7) pi-OED DoE

Up to this point, we have given the experiment ξ_{LH} with its experimental results $(y_1, \ldots, y_5)$ and estimated the initial parameter θ_{init} from it. To reduce the uncertainty of the estimation of the first parameter component θ_0, we conduct the pi-OED calculation with five new experimental designs $x_6, \ldots, x_{10}$ granted. This corresponds to finding a global solution of the minimization problem

$$\underset{x_6,\ldots,x_{10}}{\operatorname{argmin}} I^{-1}(\hat{\theta}, (x_1, \ldots, x_{10}))_{11}.$$

We solve that minimization problem by applying the differential evolution algorithm described above to the objective function

$$(z_1, \ldots, z_{10}) \mapsto I^{-1}(\hat{\theta}, (x_1, \ldots, x_5, (z_1, z_2), \ldots, (z_9, z_{10})))_{11}$$

with search space

$$(T_l, T_u) \times (SoC_l, SoC_u) \times \cdots \times (T_l, T_u) \times (SoC_l, SoC_u).$$

We thereby obtain some $z = (z_1, \ldots, z_{10})$. We then extract the pi-OED $\xi_{pi} = (x_6, \ldots, x_{10})$ from z by setting

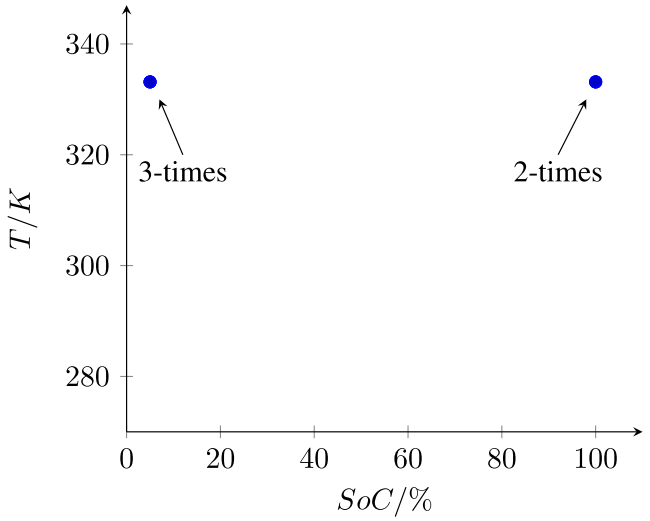


FIGURE 4. pi-OED DoE.

$x_6 = (z_1, z_2), \ldots, x_{10} = (z_9, z_{10})$. By construction, $x_6, \ldots, x_{10}$ solves

$$\operatorname*{argmin}_{x_6,\ldots,x_{10}} I^{-1}(\hat{\theta}, (x_1, \ldots, x_{10}))_{11}.$$

Figure 4 shows the resulting new pi-OED experimental plan for designs $\xi_{pi} = (x_6, \ldots, x_{10})$. Number of repetitions is indicated within the graph. Adding the pi-OED calculated new experiment $\xi_{pi} = (x_6, \ldots, x_{10})$ to the already existing ξ_{LH}, the new CRLB calculated at θ_{init} is then given by

$$I^{-1}(\theta_{init}, (x_1, \ldots, x_{10})) = \begin{pmatrix} 4.1\text{e}{-03} & -5.0\text{e}{-02} & -6.2\text{e}{-07} \\ -5.0\text{e}{-02} & 8.1\text{e}{+02} & 2.1\text{e}{-01} \\ -6.2\text{e}{-07} & 2.1\text{e}{-01} & 6.8\text{e}{-05} \end{pmatrix}.$$

This approximates the expected relative standard deviations (prior executing the new experimental designs).

8) 2nd RUN OF EXPERIMENT AND MODEL CALIBRATION

After execution of the above calculated pi-OED experiment with 5 experimental designs (yielding 5 times 6 additional data points), the corresponding statistical model reflecting the joint experiments of LH-DoE and pi-OED $\xi = (x_1, \ldots, x_{10})$ is given by

$$(\mathbb{R}^{60}, \mathcal{B}^{60}, \mathcal{N}((f_\theta(T_i, SoC_i, t))_{i=1,\ldots,10}, \sigma^2 \mathbb{1}_{60}) : \theta \in \Theta).$$

We argue by analogy with step 4) that its MLE exists.

Calculating the maximum likelihood estimation yields the optimized parameter estimation

$$\begin{aligned} \theta_{opt} &= \operatorname*{argmin}_{\theta} \sum_{i=1}^{10} \sum_{j=0}^{5} (y_{i,j} - f_\theta(T_i, SoC_i, t))^2 \\ &= (3.93, 2344.44, 0.81). \end{aligned}$$

Thereby, parameter θ_{opt} is calculated by applying the differential evolution optimizer described above to the objective function

$$\theta \mapsto \sum_{i=1}^{10} \sum_{j=0}^{5} (y_{i,t} - f_\theta(T_i, SoC_i, t_j))^2$$

where the search space is set to Θ.

Again, bootstrapping with $N_{rep} = 1000$ fold resampling within our (now 60-dimensional) sample space allows us to quantify the statistical distribution of our parameter estimator. Figures 3(d)-(f) show the normalized distributions of the estimated parameter vector component distributions after pi-DOE as obtained via bootstrapping. The numbers are scaled evenly with the *true* parameter value as the center and a range of ±40% around this value. The probability density function (pdf) of normal distributions with mean values defined by the true parameter values θ_i and variances by respective diagonal entries of the CRLB is shown in red within the according sub-figures.

TABLE 2. Relative expected standard deviation of θ_{opt}.

$\theta_{opt,i}$	θ_0	θ_1	θ_2
$\sigma_{rel,i}$	1.6%	1.2%	1.0%

9) 2nd MODEL EVALUATION

The calculated LOOCV MAE after pi-OED and computer experimental run is 0.0022. Observe that this error is again in the range of the known measurement error $\sigma = 0.002$. The determinant of the corresponding FIM at θ_{opt} turns out to be greater than zero yielding a CRLB at

$$I^{-1}(\theta_{opt}, (x_1, \ldots, x_{10})) = \begin{pmatrix} 4.2\text{e}{-03} & -5.5\text{e}{-02} & -7.1\text{e}{-07} \\ -5.5\text{e}{-02} & 8.2\text{e}{+02} & 2.1\text{e}{-01} \\ -7.1\text{e}{-07} & 2.1\text{e}{-01} & 6.8\text{e}{-05} \end{pmatrix}.$$

Accordingly, the relative expected standard deviation for each parameter component $\theta_{opt,i}$ with respect to the optimized parameter estimate θ_{opt} is shown in Table 2.

10) PREDICTION QUALITY SATISFACTORY?

Since all approximate relative standard deviations are below five percent, we accept the parameter quality. Since the LOOCV MAE is in the range of the measurement error we accept the model quality and end our experiments.

B. BENCHMARKING

In this paper, we claim our proposed pi-OED methodology effectively and efficiently minimizes a chosen individual parameter's variance supporting an unbiased estimator approximating the true model parameter. Our main objects of interest therefore are the

1) mean of the parameter estimations and
2) variance of individual parameter (component) estimations.

TABLE 3. Benchmark of several DoE methods. Bold values represent the lowest value in the respective column.

	i=0					i=1					i=2				
DoE	M_i	$M_{rel,i}$	$\sqrt{V_i}$	$\hat{\sigma}_i$	$\hat{\sigma}_{rel,i}$	M_i	$M_{rel,i}$	$\sqrt{V_i}$	$\hat{\sigma}_i$	$\hat{\sigma}_{rel,i}$	M_i	$M_{rel,i}$	$\sqrt{V_i}$	$\hat{\sigma}_i$	$\hat{\sigma}_{rel,i}$
Random	4.001	**0.0**	0.112	0.113	0.6	2300.132	**0.0**	42.629	41.876	-1.8	0.8	0.0	0.009	0.009	-3.3
LH	4.013	0.3	0.184	0.179	-2.8	2300.646	0.0	38.109	37.398	-1.9	0.8	0.0	**0.008**	**0.008**	-2.0
pi	4.002	0.0	**0.067**	**0.067**	-0.4	2298.752	-0.1	28.966	28.381	-2.0	0.8	-0.1	0.008	0.008	-4.0
D-opt	3.999	-0.0	0.072	0.074	2.7	2300.222	0.0	**27.054**	**26.617**	-1.6	0.8	**0.0**	0.008	0.008	-3.0

In addition to the theoretical guarantees given in II, we provide a benchmarking of pi-OED in order to strengthen our claim. Specifically, we compare the (estimated) mean and variances of the above computer experiment with those of three other computer experiments as calculated by

1) **Random Sampling**: We draw 10 experimental designs uniformly distributed over X.
2) **Latin-Hypercube Sampling**: We calculate a Latin-Hypercube experiment with 10 experimental designs.
3) **D-optimal DoE**: We replace the pi-OED in the above workflow with the D-optimal design of experiments (maximizing the FIM's determinant) proposed by Atkinson and Donev [9] at θ_{init} with five experimental designs.

For each of the above defined computer experiments, we bootstrap $N = 1000$ experimental evaluations to calculate their according parameter estimate $\hat{\theta}^j (j = 1, \ldots, N)$.

We estimate the mean of the parameter estimator by

$$M = \frac{1}{N} \sum_{j=1}^{N} \hat{\theta}^j \tag{33}$$

and the variance of the parameter component estimators $\hat{\theta}_i^j$ by

$$V_i = \frac{1}{N-1} \sum_{j=1}^{N} (M_j - \hat{\theta}_i^j)^2 \tag{34}$$

for $i = 0, 1, 2$. Note that both are unbiased estimators.

For each computer experiment we compare its parameter estimators'

1) estimated mean M.
2) relative deviation from the true parameter value

$$M_{rel,i} = \frac{M_i - \theta_{true,i}}{\theta_{true,i}} \tag{35}$$

in percent.

3) estimated standard deviations $\sqrt{V_i}$.
4) approximate standard deviation at true parameter θ_{true}

$$\hat{\sigma}_i = \sqrt{I^{-1}(\theta_{true})_{ii}}. \tag{36}$$

5) relative deviation of approximate standard deviation at true parameter from the estimated standard deviation

$$\hat{\sigma}_{rel,i} = \frac{\hat{\sigma}_i - \sqrt{V_j}}{\sqrt{V_j}} \tag{37}$$

in percent.

The benchmarking results are shown in Table 3. Recall the true parameter to be $\theta_{true} = (4, 2300, 0.8)$.

IV. DISCUSSION

Based on theory of section II and our example case in section III, the pi-OED methodology demonstrates a) to provide an experimenter with information about achievable parameter extraction quality based on an experimental plan prior starting the experiments plus b) to enable calculation of an experimental plan for improved estimation of individually selectable parameter components. As experiments are subject to errors, also the estimation of a model parameter vector θ is subject to an experimental error related distribution. Applying a bootstrapping approach to computer experiments allowed us to a) quantify the variance of the estimation of θ, and b) demonstrate its components to approximately approach their respective diagonal entry of the inverse FIM (i.e. CRLB). In the context of our example, the newly proposed workflow was run through twice (see Figure 3). The first iteration was executed with an experimental plan according LH sampling, the second experiment was planned according pi-OED minimizing the above mentioned CRLB. Both runs successfully confirmed our claims:

1) The achievable quality of the model parameter estimates has been correctly predicted before the associated experiment was performed.
2) pi-OED allowed to improve individual parameter estimates drawn from a given number of experiments for a specifically selected parameter.

In our example, we defined a 5% confidence level with respect to the relative parameter estimate variance for model and parameter acceptance. After the first (LH based DoE) run, estimation quality of one parameter component (θ_0) missed this target and was chosen to be individually improved. The pi-OED based second iteration of experiments surpassed the 5% level. Model validation itself was

performed using a LOOCV approach with MAE as error measure. Model validation, however, can be misleading as long as model parameter estimations are not precise enough. In this case, insufficient prediction quality cannot be assigned to inadequate model or parameter estimation quality. Our pi-OED workflow requests a calibrated model to meet simultaneously quality criteria for both, the overall model *and* parameter quality. Only in case MAE of the LOOCV *and* parameter estimation quality are meeting the confidence criterion, we may accept our model as calibrated within a given confidence level.

Based on an individual experiment, variance of a parameter estimation cannot be estimated. On the other hand, our chosen bootstrapping approach for quantification of parameter estimation variance is only feasible for computer experiments. However, as demonstrated above CRLB proved to be a valid quality measure of the parameter estimation for any experimental plan. Experimentally (bootstrapping based) derived distributions reflect their theoretical distribution qualitatively (shape) and quantitatively (mean and variance). The results, in total, thereby demonstrate the capability of our chosen methodological approach.

When comparing the results of several methods for designing experiments, pi-OED is equal or outperforms the random sampling and LH sampling in all benchmarking metrics for all parameters (see Table 3). Compared to D-optimal design, pi-OED leads to a slightly improved standard deviation of the individually chosen model parameter. This is not surprising since the D-optimal design minimizes the FIM determinant in total which is, however, dominated by maximum individual (co)variance contributions. Accordingly for our example, the standard deviation of remaining parameters is lower with the D-optimal design while being slightly above that of our individually selected parameter.

The experimental (see III-A) and benchmarking (see III-B) results for our pi-OED methodology as summarized in Table 3 reflect:

1) The estimated mean based on experimental data is sufficiently close to the real parameter value.
2) Variances assigned to estimated parameter values based on experimental data are sufficiently close to their approximations by CRLB entries.
3) pi-OED enables experimenters to calculate experimental plans allowing to improve estimation quality for any individually selectable parameter.
4) pi-OED outperforms state of the art designs of experiments with respect to individually chosen parameters.

V. CONCLUSION

Parametric models can be calibrated using experimental results. To allow this in an environment of expensive experiments, an efficient and effective DoE is essential. The novel pi-OED methodology enables experimenters to calculate an optimal experimental design improving the quality of a parametric model and individually selectable parameters. It is universally applicable to all parametric models. The exemplary application of the pi-OED workflow on the parameter determination of a battery aging model in a simulation-based environment demonstrated our workflow and quantified our claims. pi-OED opens up a wide range of opportunities. In a next step, we intend to extend the current single-criteria approach to a multi-objective optimization. This would allow experimenters to identify Pareto-optimal trade-offs of experimental plans with respect to a multitude of experimental target indicators such a time and cost of an experiment.

APPENDIX
PROOFS

Proof of Corollary 7: Since

$$C_\theta(T) - I^{-1}(\theta) \tag{38}$$

is positive semi-definite, we obtain

$$\mathrm{var}_\theta(T_i) - I^{-1}(\theta)_{ii} = e_i^T(C_\theta(T) - I^{-1}(\theta))e_i \geq 0.$$

This proves the claim. □

Proof of Proposition 9: If an efficient estimator g exists, then, g satisfies

$$\frac{\partial}{\partial\theta}\ln(\rho_\theta(x) = I(\theta)(g(x) - \theta)) \tag{39}$$

by theorem 6. Let T be a MLE. Maximizing

$$\rho(-, x) : \Theta \to \mathbb{R}, \theta \mapsto \rho_\theta(x)$$

is equivalent to maximizing $\ln\rho(-, x)$ because ln is monotonically increasing. In particular,

$$0 = \frac{\partial}{\partial\theta}\ln(\rho(T(x), x)) = I(T(x))(g(x) - T(x)). \tag{40}$$

Note that we use that the MLE exists on the open subset $\Theta \subset \mathbb{R}^n$. Since $I(T(x))$ is invertible by assumption, we deduce $g(x) = T(x)$ and, in particular, T to be efficient. □

Proof of Proposition 12: Denote by $I'(\theta)$ the FIM corresponding to $(\mathbb{R}^{n'}, \mathcal{B}^{n'}, P'_\theta : \theta \in \Theta)$. Then, $I'(\theta)$ and $I(\theta)$ are positive semi-definite by proposition 11 and $I(\theta)$ is positive definite (i.e. invertible) by assumption. Since the density function of $P_\theta \otimes P'_\theta$ is given by the product of density functions of P_θ and P'_θ, we calculate

$$\tilde{I}(\theta) = I(\theta) + I'(\theta). \tag{41}$$

Recall the sum of a positive definite and a positive semi-definite matrix to be positive definite. Thus, $\tilde{I}(\theta)$ is positive definite. Furthermore,

$$\tilde{I}(\theta) - I(\theta) = I'(\theta)$$

is positive semi-definite. Thus,

$$I^{-1}(\theta) - \tilde{I}^{-1}(\theta)$$

is positive semi-definite. We obtain

$$I^{-1}(\theta)_{ii} - \tilde{I}^{-1}(\theta)_{ii} = e_i^T (I^{-1}(\theta) - \tilde{I}^{-1}(\theta)) e_i \geq 0 \tag{42}$$

for all i. □

Proof of Proposition 13: The FIM is given by the Gram matrix

$$I(\theta) = C^T C \tag{43}$$

for

$$C := (\frac{\partial}{\partial \theta_i} f_\theta(x_l))_{i=1,\dots,m, l=1,\dots,N} \tag{44}$$

by example 5. Then, $I(\theta)$ is invertible if and only if the column vectors of C are linearly independent if and only if there exists a basis of $\mathbb{R}^m$ consisting of column vectors of C. The column vectors of C are given by

$$C_l = (\frac{\partial}{\partial \theta_i} f_\theta(x_l))_{i=1,\dots,m} \tag{45}$$

for $l = 1, \dots, N$. Without loss of generality, we assume

$$C_1, \dots, C_m$$

to be a basis of $\mathbb{R}^m$. We recognize those to be the row vectors of $DF(\theta)$, where DF is the total derivative of F. Thus, those are a basis of $\mathbb{R}^m$ if and only if $DF(\theta)$ is invertible. By the inverse function theorem we obtain the claim. □

Proof of Proposition 14: Since Θ is bounded so is $\bar{\Theta}$. In particular, $\bar{\Theta}$ is bounded and closed, hence, quasi-compact. Recall continuous, real valued maps from quasi-compact topological spaces to attain their maximum and minimum. In particular, the continuous map $\bar{\rho}(-, x)$ attains its maximum for all $x \in X$. □

VI. ACKNOWLEDGMENT

(Nicolai Palm and Florian Stroebl contributed equally to this work.)

REFERENCES

[1] H. Lauschmann and V. S. Beneš, "Spatial statistics in the material research," in *Industrial Statistics*, C. P. Kitsos and L. Edler, Eds. Heidelberg, Germany: Physica-Verlag HD, 1997, pp. 285–293.

[2] M. J. Anderson, L. Liao, and H. C. Basoalto, "An assessment of statistical models of competitive growth during transient ostwald ripening in turbine disc nickel-based superalloys," *Model. Simul. Mater. Sci. Eng.*, vol. 30, no. 7, Oct. 2022, Art. no. 075005.

[3] J. Pinheiro, B. Bornkamp, E. Glimm, and F. Bretz, "Model-based dose finding under model uncertainty using general parametric models," *Statist. Med.*, vol. 33, no. 10, pp. 1646–1661, May 2014.

[4] J. Heitzmann, Y. Thoma, R. Bricca, M.-C. Gagnieu, V. Leclerc, S. Roux, A. Conrad, T. Ferry, and S. Goutelle, "Implementation and comparison of two pharmacometric tools for model-based therapeutic drug monitoring and precision dosing of daptomycin," *Pharmaceutics*, vol. 14, no. 1, p. 114, Jan. 2022.

[5] S. M. Afonin, "Structural–parametric model electroelastic actuator nano- and microdisplacement of mechatronics systems for nanotechnology and ecology research," *MOJ Ecol. Environ. Sci.*, vol. 3, no. 5, Sep. 2018.

[6] C. Tamm and S. Perfetto, "Design and optimization of mechatronic systems using a holistic and parametric simulation approach," *IFAC-PapersOnLine*, vol. 52, no. 15, pp. 271–276, 2019.

[7] R. A. Fisher, "The arrangement of field experiments," *J. Ministry Agricult.*, no. 33, pp. 503–513, 1926, doi: 10.23637/rothamsted.8v61q.

[8] R. A. Fisher, *The Design of Experiments*. New York, NY, USA: Hafner Press, 1974.

[9] A. C. Atkinson and A. N. Donev, *Optimum Experimental Designs* (Oxford Statistical Science Series), no. 8. Oxford, U.K.: Oxford Univ. Press, 1992.

[10] G. Franceschini and S. Macchietto, "Model-based design of experiments for parameter precision: State of the art," *Chem. Eng. Sci.*, vol. 63, no. 19, pp. 4846–4872, Oct. 2008.

[11] V. Fedorov, "Optimal experimental design: Optimal experimental design," *WIREs Comp Stat*, vol. 2, no. 5, pp. 581–589, 1972.

[12] P. Reisenthel and D. Lesieutre, "A numerical experiment on allocating resources between design of experiment samples and surrogate-based optimization infills," in *Proc. 52nd AIAA/ASME/ASCE/AHS/ASC Struct., Struct. Dyn. Mater. Conf.*, Denver, CO, USA, Apr. 2011, pp. 1–7.

[13] S. M. Kay, *Fundamentals of Statistical Signal Processing* (Signal Processing Series). Englewood Cliffs, NJ, USA: Prentice-Hall, 1993.

[14] A. Charnes, E. L. Frome, and P. L. Yu, "The equivalence of generalized least squares and maximum likelihood estimates in the exponential family," *J. Amer. Statist. Assoc.*, vol. 71, pp. 169–171, 1976.

[15] H.-O. Georgii, *Stochastik: Einführung die Wahrscheinlichkeitstheorie und Statistik* (De Gruyter Studium), 5th ed. Berlin, Germany: De Gruyter, 2015.

[16] S. S. Shapiro and M. B. Wilk, "An analysis of variance test for normality (complete samples)," *Biometrika*, vol. 52, nos. 3–4, pp. 591–611, Dec. 1965.

[17] M. D. McKay, R. J. Beckman, and W. J. Conover, "A comparison of three methods for selecting values of input variables in the analysis of output from a computer code," *Technometrics*, vol. 21, no. 2, p. 239, May 1979.

[18] T. Fushiki, "Estimation of prediction error by using K-fold cross-validation," *Statist. Comput.*, vol. 21, no. 2, pp. 137–146, Apr. 2011.

[19] J. D. Rodriguez, A. Perez, and J. A. Lozano, "Sensitivity analysis of K-fold cross validation in prediction error estimation," *IEEE Trans. Pattern Anal. Mach. Intell.*, vol. 32, no. 3, pp. 569–575, Mar. 2010.

[20] J. Li, "Assessing the accuracy of predictive models for numerical data: Not r nor r^2, why not? Then what?" *PLoS ONE*, vol. 12, no. 8, Aug. 2017, Art. no. e0183250.

[21] C. Tofallis, "A better measure of relative prediction accuracy for model selection and model estimation," *J. Oper. Res. Soc.*, vol. 66, no. 8, pp. 1352–1362, Aug. 2015.

[22] S. Park, D. Kato, Z. Gima, R. Klein, and S. Moura, "Optimal experimental design for parameterization of an electrochemical lithium-ion battery model," *J. Electrochem. Soc.*, vol. 165, no. 7, pp. A1309–A1323, 2018.

[23] D. N. How, M. A. Hannan, M. H. Lipu, and P. J. Ker, "State of charge estimation for lithium-ion batteries using model-based and data-driven methods: A review," *IEEE Access*, vol. 7, pp. 136116–136136, 2019.

[24] B. Efron and R. Tibshirani, "The bootstrap method for assessing statistical accuracy," *Behaviormetrika*, vol. 12, no. 17, pp. 1–35, Jan. 1985.

[25] J. Schmalstieg, S. Käbitz, M. Ecker, and D. U. Sauer, "A holistic aging model for Li(NiMnCo)O2 based 18650 lithium-ion batteries," *J. Power Sources*, vol. 257, pp. 325–334, Jul. 2014.

[26] M. Naumann, M. Schimpe, P. Keil, H. C. Hesse, and A. Jossen, "Analysis and modeling of calendar aging of a commercial $LiFePO_4$/graphite cell," *J. Energy Storage*, vol. 17, pp. 153–169, Jun. 2018.

[27] M. Naumann, F. B. Spingler, and A. Jossen, "Analysis and modeling of cycle aging of a commercial $LiFePO_4$/graphite cell," *J. Power Sources*, vol. 451, Mar. 2020, Art. no. 227666.

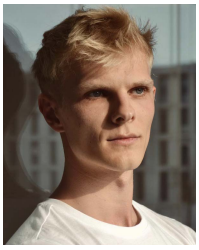
[28] M. Mühlbauer, F. Rang, H. Palm, O. Bohlen, and M. A. Danzer, "Pareto-optimal power flow control in heterogeneous battery energy storage systems," *J. Energy Storage*, vol. 48, Apr. 2022, Art. no. 103803.

[29] R. Storn and K. Price, "Differential evolution—A simple and efficient heuristic for global optimization over continuous spaces," *J. Global Optim.*, vol. 11, no. 4, pp. 341–359, Dec. 1997.

[30] P. Virtanen et al., "SciPy 1.0: Fundamental algorithms for scientific computing in Python," *Nature Methods*, vol. 17, no. 3, pp. 261–272, Mar. 2020.

[31] G. I. Webb et al., J.-A. Ting, S. Vijayakumar, S. Schaal, and L. D. Raedt, "Leave-one-out cross-validation," in *Encyclopedia of Machine Learning*, C. Sammut and G. I. Webb, Eds. Boston, MA, USA: Springer, 2011, pp. 600–601.

[32] F. Pedregosa, G. Varoquaux, A. Gramfort, V. Michel, B. Thirion, O. Grisel, M. Blondel, P. Prettenhofer, R. Weiss, and V. Dubourg, "Scikit-learn: Machine learning in Python," *J. Mach. Learn. Res.*, vol. 12, pp. 2825–2830, Nov. 2011.



NICOLAI PALM received the B.Sc. and M.Sc. degrees in mathematics from the University of Regensburg, Germany, in 2018 and 2021, respectively. He is currently pursuing the doctoral degree with Ludwig Maximilian University (LMU), Munich, Germany. He works as a Data Scientist for startups and is involved in AI-based industrial projects. His research interests include optimal design of experiments and multi-criteria optimization approaches.

FLORIAN STROEBL received the B.Eng. degree in electrical engineering and the M.Eng. degree in electrical and microsystems engineering from the University of Applied Sciences (UAS) OTH Regensburg, Germany, in 2019 and 2020, respectively. In 2021, he joined as a Research Assistant with the Institute for Sustainable Energy Systems, Munich, Germany, as part of a publicly funded research program. His special interests include the optimization of highly cost intensive and time consuming experiments with application focus on lithium ion battery cell degradation for aging analysis.

HERBERT PALM received the Ph.D. degree in physics, in 1994. He worked in leading international Research and Development and operations positions of both high-tech start-ups and corporate companies until 2008. In 2008, he took over a full-time professorship at the University of Applied Sciences Munich (UAS Munich). Since 2014, he has been the Head of the UAS Munich Systems Engineering Master's program. He calls VUCA environment his home. From an application point of view, he focuses on the support of projects with a sustainable energy system link. Within this context, he is one of the founding members of the Institute of Sustainable Energy Systems (ISES) in Munich. His research interests include analysis, design, and optimization of complex systems. He is a Senior Member of INCOSE and the German Physical Society (DPG).