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Efficient parameter estimation for spectral sensor data by a linear transformation

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In this contribution we introduce and evaluate a new approach for solving inverse problems in proximity of a working point with very low computational effort. The non-linear, multi-parametric, complex function will be approximated and inverted by a set of decoupled single parametric, linear equations originating from a sensitivity analysis. The used linear projection condenses the knowledge of the transfer characteristic of the system and provides an alternative to model based and look-up table approaches. The fast estimation of multiple parameters in a limited parameter range is suitable for control applications or investigation of aging and other degeneration processes.

Keywords: Impedance Spectroscopy; Linear Transformation; Parameter Estimation

INTRODUCTION

To increase the amount of information that can be obtained in a single measurement, multi-spectral measurement techniques have been introduced in the recent decades. Multiple influences on a sensor signal can be separated on a frequency scale due to the fact that different effects or mechanisms that sum up to the sensor signal act in different frequency ranges. Common approaches for data analysis of such multispectral data involve a model for regression and a nonlinear optimization process [1]. In many cases the optimization is done by an iterative algorithm [2]. The repeated calculation of the model and the evaluation of some loss functions consume a large amount of computational resources and time. For embedded system solutions, high demands on the dynamics of the measurement and data evaluation speed or models that require high computational effort in the nonlinear regression make the classical approaches unsuitable [3].

The introduced approach is inspired by the Tasselled-Cap-algorithm [4], which is projecting spectral information into a new subspace where quantities of interest are linear independent. The Gram-Schmidt orthogonalization within the Tasseled-Cap-algorithm performs a compensation of independent components in the data set to project the data in subspace. This projection is decoupling the influence by compensation of cross-sensitivities of desired quantities [5].The orthogonalization uses a pre-defined set of vectors as basis for the new subspace. The possibility to choose these basis vectors based on the sensitivity of the different quantities of interest, is a useful property in the applications with inverse problems. In the decoupled subspace the desired quantity is then obtained by solving the resulting linear system. The projection formula condenses the knowledge on the systems transfer characteristic in a certain working point and is used to estimate the quantity of interest instead of using models or look up tables. Instead of using the full spectral data the projection uses only two data points selected in a way that they contain maximum information on the quantity of interest. With the presented conditions for these points, they can be chosen in an automated and objective way. The limitation of the measurement to these selected frequency points reduces the measurement time and hardware requirements.

LINEAR PROJECTION

Usually the transfer characteristics of a sensor are described by some model. This model is defined by a set of parameters x. The goal now is to derive the model parameters based only on the measurements performed with the sensor. This is called an inverse problem and usually the calculation of the parameters is challenging as noise in the measurement makes the inversion process ill-posed. With a sensor application in mind one of these parameters is usually of interest and is therefore called a measurement quantity or measurand.

Let us consider that the measurement is performed in some working point (WP) of the sensor. In the proximity of a WP any continuously differentiable function Z characterized by a parameter set x_1 to x_m can be expressed as linear approximation $\tilde{Z}$:

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$$\tilde{Z} = \sum_{k=1}^{m} \frac{\partial Z}{\partial x_k} \Delta x_k + Z_{WP} \,, \qquad (1)$$

This approximation consists of m products of the sensitivity with respect to the parameters and their corresponding distance from the working point Δx_k and one constant term as working point offset Z_{WP}.In the next step a weighted sum of n of these approximations with weighting factors from a_1 to a_p can be made and rearranged to obtain equation (2). The expression on the right side consists now of *m* weighted sums of (1). In the case of spectral data each of the *p* used approximations represents one measurement at a certain frequency:

$$\sum_{n=1}^{p} a_n \tilde{Z}_n = \sum_{k=1}^{m} \left(\left[\sum_{n=1}^{p} \frac{a_n \partial Z_n}{\partial x_k} \right] \Delta x_k \right) + \sum_{n=1}^{p} a_n Z_{WP,n} \,, \qquad (2)$$

The sensitivity of the entire sum of linear approximations is defined by a suitable set of weighting factors a_n. To obtain the weights a_n we have to introduce a constraint on the sensitivity. In most sensor applications it is useful to set the sensitivity of the wanted parameter x_n (e.g. x_1) to the non-zero value S_1 and all other to zero:

$$\begin{aligned} \sum_{\mathrm{n=1}}^{\mathrm{p}} \mathrm{a_n} \frac{\partial \mathrm{Z_n}}{\partial \mathrm{x_1}} &= \mathrm{S_1} \\ \sum_{\mathrm{n=1}}^{\mathrm{p}} \mathrm{a_n} \frac{\partial \mathrm{Z_n}}{\partial \mathrm{x_2}} &= 0 \vdots, \\ \sum_{n=1}^{p} a_n \frac{\partial Z_n}{\partial x_m} &= 0. \end{aligned} \qquad (3)$$

For the case of spectral data solving the linear system of equations (3) will result in a sum of one quantity at different frequencies, which is now depending on only one parameter:

$$\sum_{n=1}^{p} a_n Z_n = S_1 (x_1 - x_{WP}) + \sum_{n=1}^{p} a_n Z_{WP,n} \,, \qquad (4)$$

Rearranging equation (4) and solving for the desired parameter gives:

$$x_{1,est} = \sum_{n=1}^{p} \frac{a_n Z_n}{S_1} + x_{offset} \,, \qquad (5)$$

The additional constant x_{offset} is calculated with the parameter value at the working point x_{WP}:

$$x_{offset} = x_{WP} - \sum_{n=1}^{p} \frac{a_n Z_{WP,n}}{S_1} \,. \qquad (6)$$

The defined sensitivity of the weighted sum to the wanted parameter S_1 in equation (3) results in a linear scaling of the weighting factors that are removed by inverting equation (4). For the reason of numerical simplicity it is recommended to choose 1 for the sensitivity.

$$S_1 = 1, \qquad (7)$$

To successfully perform this procedure, the sensitivity of the desired quantity must be high and the difference to the sensitivity of other system parameters must be maximal. In the simplest case of *p=m* the parameter vector of all used frequencies needs to be linear independent. This property has to be ensured by the selection of used frequency points like it will be demonstrated. The analyzed transfer characteristic can be any frequency dependent transfer parameter like gain, phase, real or imaginary part. Also a mixed support of those quantities, as two linked real quantities, may be possible if it is required by the application [5]. A generalization for complex transfer functions by the use of complex weighting factors might be a future improvement of the approach. The used quantity as base for the algorithm in the example is the imaginary part of the impedance as real scalar value. For the estimation of other parameters similar formulas like equation (5) can be obtained by adjusting non-zero sensitivity on the right hand side of the system of equations (3).

DEMOSTATION FOR ONE ESTIMATED PRAMETER

Generic model

In this section, the introduced algorithm is tested and evaluated using generic data to avoid uncertainties of the measurement process. The data is generated from a model for the complex impedance of a solid state electrolyte including electrode effects. The electrodes are represented by the serial resistance R_s, the parallel resistance R_p and the electrode capacitance C_1. The ionic contribution is represented by the Warburg-impedance $\underline{Z}_\mathrm{w}$ with reflecting boundary condition [6]:

$$\underline{Z} = R_s + \frac{1}{j\omega C_1 + \frac{1}{R_p} + \frac{1}{\underline{Z_W}}} \,, \qquad (8)$$

$$\underline{Z_W} = \frac{RT}{I^2 F^2 c_x A \sqrt{2\omega D}} (1 - j) \coth\left(\delta (1 + j) \sqrt{\frac{\omega}{2D}} \right), \qquad (9)$$

The aim of the data analysis is to estimate the ion concentration c_x while the diffusion coefficient D is unknown. The structure of equation (9) ensures that parts of the impedance spectra have different sensitivity with respect to both unknown parameters. The working point is chosen according table I but it does not refer to a special application. The model represents a general problem in

impedance spectroscopy, when a systematic behavior to the parameters like in figure 1is observed and a multi parametric interpretation is needed.

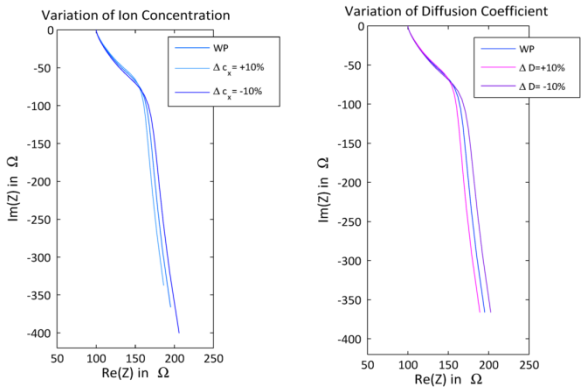

Fig. 1. Transfer characteristic of the generic model due to 10% variation of the two non-constant parameters.

Table 1. Working Point Model Paramters

Name	*Symbol*	*Value*
Serial Resistor	R_s	100Ω
Capacitance of the Electrodes	C_1	$500 * 10^{-9}\mathrm{F}$
Paralel Resistance	R_p	$5\ \mathrm{k}\Omega$
Universal Gas Constant	R	$8{,}31447\,\frac{\mathrm{J}}{\mathrm{mol\,K}}$
Faraday Constant	F	$96485{,}34\,\frac{\mathrm{C}}{\mathrm{mol}}$
Distance of Electrodes	$\boldsymbol{\delta}$	$10^{-6}\mathrm{m}$
Elecrtrode Area	A	$10^{-4}\mathrm{m}^2$
Charge per Ion	I	1
Temperature	T	298 K
Ion Concentration WP	c_x	$10^{-3}\,\frac{\mathrm{mol}}{\mathrm{m}^3}$
Diffusion Coefficient WP	D	$10^{-9}\,\frac{\mathrm{m}^2}{s}$

Selection of data support

The first step is the selection of a suitable spectral representation of the system response. Since some representations are more sensitive to certain physical effects this selection is based on the knowledge of the investigated physical system. In this example the targeted effect is the frequency dependent capacitance of the double layer which is represented by the imaginary part of the impedance in equation (8). Due to the reduced influence of the resistive effects caused by the serial and parallel resistance, the entire data analysis focuses exclusively on the imaginary part of the impedance of the entire system.

To estimate the ion concentration in a set of two unknown parameters at least two points in the spectrum of the imaginary part are needed as data support. These points must have a strong sensitivity to the parameter of interest and a linear independent sensitivity vector to solve (3). The presence of a high absolute value of sensitivity is represented by the first criterion K_I for each possible combination of the spectral data at frequencies index *d* and *e*:

$$K_I = \left|\frac{\partial Z_d}{\partial x_1}\right| + \left|\frac{\partial Z_e}{\partial x_1}\right|, \tag{10}$$

The independence of the sensitivity information is checked by the second criterion K_{II} in the form of a normalized difference of the sensitivity with respect to the other parameter:

$$K_{II} = \frac{\partial Z_d}{\partial x_2}\frac{\partial x_1}{\partial Z_d} - \frac{\partial Z_e}{\partial x_2}\frac{\partial x_1}{\partial Z_e}, \tag{11}$$

Both criteria have a high absolute value for suitable combinations and tend to zero for unsuited combinations. This numerical property can be used to combine both criteria to one common criterion by multiplication:

$$K_{ges} = K_I K_{II} = \left(\frac{\partial Z_d}{\partial x_2}\frac{\partial x_1}{\partial Z_d} - \frac{\partial Z_e}{\partial x_2}\frac{\partial x_1}{\partial Z_e}\right)\left(\left|\frac{\partial Z_d}{\partial x_1}\right| + \left|\frac{\partial Z_e}{\partial x_1}\right|\right), \tag{12}$$

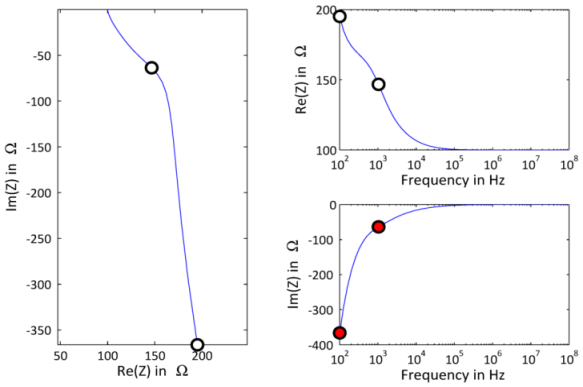

Fig. 2. Selected data support in the spectrum at working point conditions

The common criterion has only a high absolute value for combinations of data from different frequencies that match both criteria. The sensitivities are obtained by numerical derivation with a variation of the parameters of ±1% in equation (13) and (14):

$$\frac{\partial Z_n}{\partial x_1} = \frac{Im\left\{\underline{Z_n}(1{,}01x_1) - \underline{Z_n}(0{,}99x_1)\right\}}{0{,}02x_1}, \tag{13}$$

$$\frac{\partial Z_n}{\partial x_2} = \frac{Im\left\{\underline{Z_n}(1{,}01x_2) - \underline{Z_n}(0{,}99x_2)\right\}}{0{,}02x_2}. \tag{14}$$

The analysis of all possible combinations in the spectrum reveals a maximum for the indices 1 and

11 at the frequencies f_1 at 100 Hz and f_2 at 1001 Hz illustrated in figure 2. The imaginary part of the impedance Z_1 and Z_2 at those frequencies is used as data support for the linear transformation to separate and estimate the parameters.

Calculation of the transformation formula

With two parameters to separate the system of equations (3) is reduced to:

$$a_1 \frac{\partial Z_1}{\partial x_1} + a_2 \frac{\partial Z_2}{\partial x_1} = 1 \frac{m^3 \Omega}{mol}, \quad (15)$$

$$a_1 \frac{\partial Z_1}{\partial x_2} + a_2 \frac{\partial Z_2}{\partial x_2} = 0 \frac{s\Omega}{m^2}. \quad (16)$$

Similar to equations (13) and (14) the required numerical derivative at the working point conditions can be obtained as:

$$\frac{\partial Z_1}{\partial x_1} = 3{,}1298 \cdot 10^5 \frac{m^3 \Omega}{mol}, \quad (17)$$

$$\frac{\partial Z_2}{\partial x_1} = 5{,}6343 \cdot 10^4 \frac{m^3 \Omega}{mol}, \quad (18)$$

$$\frac{\partial Z_1}{\partial x_2} = -1{,}0913 \cdot 10^3 \frac{s\Omega}{m^2}, \quad (19)$$

$$\frac{\partial Z_2}{\partial x_2} = 3{,}4387 \cdot 10^4 \frac{s\Omega}{m^2}, \quad (20)$$

Solving the system of equations (15, 16) results in the weighting factors $a_1 = 3{,}1769 \cdot 10^{-6}$ and $a_2 = 1{,}0082 \cdot 10^{-7}$.

To estimate the ion concentration $c_{x,est}$ the weighting factor a_1 has to be multiplied with the imaginary part of the impedance at 100 Hz and added to the imaginary part of the impedance at 1001 Hz multiplied with the weighting factor a_2. The correct offset value can be calculated with equation 6.

$$x_{1,est} = c_{x,est} = 3{,}1769 \cdot 10^{-6} \frac{mol}{m^3 \Omega} \cdot Im\{\underline{Z_1}\} + \cdots 1{,}0082 \cdot 10^{-7} \frac{mol}{m^3 \Omega} \cdot Im\{\underline{Z_2}\} - 1{,}6973 \cdot 10^{-4} \frac{mol}{m^3} \quad (21)$$

EVALUATION AND DISCUSSION

The evaluation of the procedure is performed by varying the ion concentration as well as diffusion coefficient by ±50 % in steps of 1% of the working point value and subsequent calculation of one spectrum for each combination of those two values. Equation 21 was used to estimate the ion concentration out of the resulting 10201 spectra. The result of all 10201 estimations is plotted in 101 graphs in figure 3. The red dashed line represents the perfect estimation of the ion concentration with no systematic error. The strong change in the diffusion coefficient is affecting the estimation in a very low degree and the 101 graphs nearly overlap each other perfectly. For a high variation of the ion concentrations the estimation error can reach large values due to the nonlinearity of the system and the linear approximation.

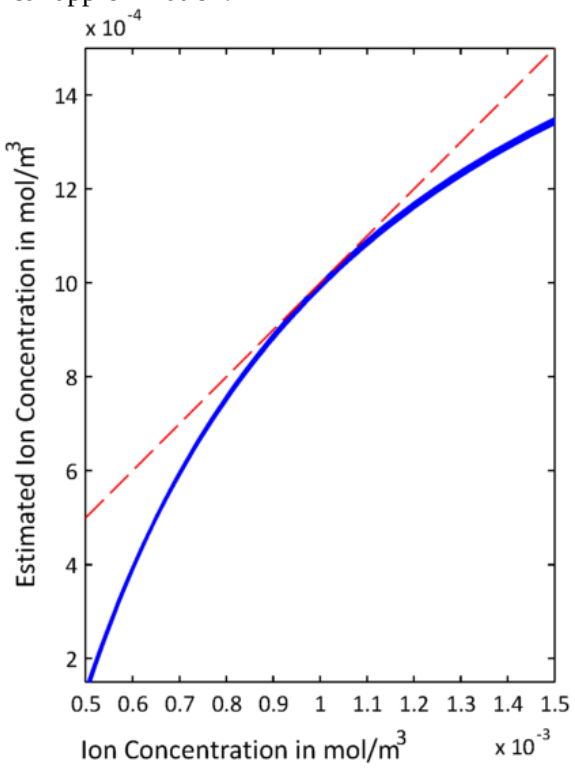

Fig. 3. Estimated ion concentration versus the true value for varying diffusion coefficients (blue graphs). An ideal estimation procedure would produce the red dashed line (red).

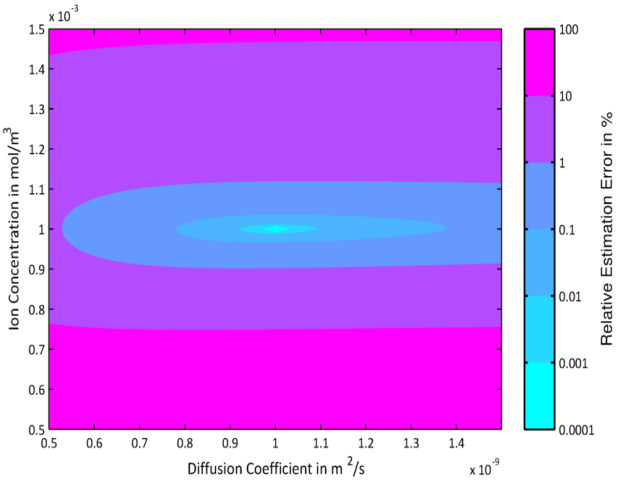

Fig. 4. Estimation error of the ion concentration relative to the actual input value.

Despite of the large error, figure 4 reveals an area with low relative estimation error in the proximity of the working point. In this area an estimation of the parameters can be performed successfully and used in control application or for tracking various aging effects.

REFERENCES

1. O. Kanoun, *Technisches Messen*, **7/8,** 73 (2006).
2. P. Büschel, U. Tröltzsch, O.Kanoun, *Electrochimica Acta*, **56**(23), 8069 (2011).
3. H.-R. Tränkler, O. Kanoun, „Paradigms for Improving Sensory Information using Multi-Sensor and Model-Based Sensor Systems“, IEEE International Conference on Systems, Signals & Devices (SSD'05), Tunisia, 2005.
4. R. J. Kauth, G. S. Thomas “The Tasselled Cap -- A Graphic Description of the Spectral-Temporal Development of Agricultural Crops as Seen by LANDSAT” LARS Symposia, 1976, p. 4B41.
5. J.G Liu, W.J Becker, T. Gerhold, W. Ricken, *Technisches Messen*, **67**(11), 484 (2000).
6. G.H.J Broers, M. Schenke, *Abhandlungen der sächsischen Akademie zu Leipzig*, **49**, 299 (1968).

ОЦЕНКА НА ЕФЕКТИВНИ ПАРАМЕТРИ НА СПЕКТРАЛНИ СЕНЗОРНИ ДАННИ ЧРЕЗ ЛИНЕЙНА ТРАНСФОРМАЦИЯ

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(Резюме)

В тази публикация ние въвеждаме и оценяваме нов подход за решаване на обратими проблеми в близост до работна точка при твърде занижени изчислителни усилия. Нелинейната, мулти-параметрична, комплексна функция ще се апроксимира и инвертира с помоща на набор от взаимно несвързани монопараметрични линейни уравнения,изведени при анализ на чувствителността. Използваната линейна проекция концентрира знанието за предавателната характеристика на трансфер и осигурява алтернативен модел, основан на подхода; вижте табличните данни. Бързата оценка на множество параметри за ограничен кръг от параметри е подходящ способ при управление на приложения или за изследване на процесите на стареене и други дегенеративни процеси.