

# Identification of fractional-order models for condition monitoring of solid-oxide fuel cell systems

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**Abstract:** With rising market deployment the condition monitoring of solid oxide fuel cell systems is gaining particular importance. The conventional approaches mainly use electrochemical impedance spectroscopy based on the repeated sinusoidal perturbation over a range of frequencies. One of the notable weaknesses of the approach is excessively long perturbation time needed to properly evaluate the impedance curve. In this paper, we propose a time-efficient approach in which, a short, persistently exciting and small-amplitude perturbation is used to excite all the relevant system eigenmodes. A model structure from a class of linear fractional order models is selected to describe the perturbed dynamics and to account for anomalous diffusion processes in the cells. Then, the model parameters are estimated directly from measured input and output records. The paper presents a computationally efficient parameter estimation procedure in which the numerical issues of differentiation of noisy signals are alleviated by using modulating functions. In practice, that means a combination of filtering and application of conventional least squares. The approach is applied on a case of health assessment of solid oxide fuel cells.

*Keywords:* Fractional order systems, time-domain identification, condition monitoring, solid oxide fuel cell systems

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## 1. INTRODUCTION

Solid oxide fuel cell systems are a promising technology for direct conversion of chemical energy bound in fuel gas into electrical energy without additional conversion steps. The high degree of fuel flexibility, high efficiency irrespective of the power scale, no need for rare materials such as platinum and lithium, make solid-oxide fuel cells (SOFCs) a perspective technology particularly suitable for stationary applications. Unfortunately, the most serious barriers to the broader commercialisation and market deployment of SOFCs are yet insufficient durability, reliability and high cost Irvine and Connor (2013).

Despite a great deal of effort dedicated to the understanding of SOFC degradation mechanisms Barelli et al. (2013), only a relatively limited set of diagnostic approaches is available. That encompasses several analytical model-based approaches Marra et al. (2016); Sorce et al. (2014), black-box approaches Sorrentino et al. (2014) and signal processing approaches Pahon et al. (2016). However, still the most frequent health assessment approach builds on the use of electrochemical impedance spectroscopy (EIS). Characteristic for EIS is that applies local perturbation to excite all the relevant dynamic modes of the system. Although it has been around for several decades, the

way it is used has not changed much. To infer on the health status, the EIS data have to be interpreted either through the change of the pattern of the EIS curve, or by interpreting changes in the parameters of the equivalent circuit models (ECM) Polverino et al. (2017); Niya et al. (2016) and distribution of relaxation times (DRT) Liu et al. (2010); Heinzmann et al. (2018). Such an approach suffers from at least two drawbacks. First, too long perturbation time is usually required to obtain high-quality EIS spectra. That means too long perturbation of the process in operating mode. Particularly critical is estimation of EIS curve at low frequencies as normally several periods of a sinusoid are required to extract precise information. Second, long perturbation times raise the chance that random disturbances come into play during the perturbation sessions, e.g. in terms of temperature drifts, variations in fuel composition. Moreover, further processing of EIS data targeting DRT and ECM in both cases requires heuristic tuning of regularisation parameters as well as exhaustive optimisation searches.

By applying excitation over a wide range of frequencies *simultaneously*, the authors in Boškoski et al. (2017) showed that the same quality of the results as in conventional EIS can be obtained at the substantially shorter perturbation times (an order of magnitude). The evaluation of EIS

curve is done by post-processing of the current and voltage signals utilizing complex wavelet transform. Apart from the much shorter perturbation session, an additional benefit is also much better resolution of the EIS curve obtained compared to the conventional EIS, as it is defined by the sampling rate. Savings in required perturbation times can gradually diminish when the required precision of the spectral reconstruction at low frequencies is increasing. To circumvent the last limitation we propose the time-domain identification of the system dynamics described by the fractional order differential equation.

The currently available approaches to fractional order systems (FOSs) identification can be split into three groups. The first one contains approaches that employ recursive linearisation techniques Sabatier et al. (2015); Malti et al. (2007). These approaches closely approximate the behaviour of a FOS within a certain frequency band. The second group comprises approaches that use algebraic manipulation of the fractional order transfer function Gehring and Rudolph (2017). The result is an integer order transfer functions whose parameters are result of an optimisation process. A challenge in all of those those approaches is its high sensitivity of the numerical derivation with noisy signals. For that purpose a class of methods making use of the modulating functions, as an elegant way to circumvent direct signal derivation, have been proposed. The idea is instead to do derivation directly on the signal to move it indirectly to the modulating function and in the closed form Eckert (2017); Liu et al. (2013); Belkhatir and Laleg-Kirati (2017); Liu and Laleg-Kirati (2015); Dai et al. (2016). Technically, a modulating function (MF) takes the form of low-pass filter. With the exception of Belkhatir and Laleg-Kirati (2017) in the most of the cases the *commensurate* transfer functions have been addressed.

In this paper we propose an algorithm for identification of a class of *incommensurate* fractional order noisy systems, which represent the best match for fuel cell system dynamics. The main results of the paper are as follows. First, a filter with an exponentially decaying MF is adopted. It serves to alleviate the problem of direct numerical derivation of the signal. Second, a parameter estimation algorithm based on a two-step nested constrained optimisation is proposed and applied to the condition monitoring of a solid oxide single-cell system. It is shown that the presumed model structure reduces the required identification time to only a fraction of the time needed for classical EIS. Finally, DRT turns to be simply a byproduct calculated through a straightforward transformation of the identified transfer function Fuoss and Kirkwood (1941). The experimental validation of the proposed methodology is performed under the degradation-free as well as the degradation-accelerated operating conditions on a SOFC.

## 2. BACKGROUND

### 2.1 Fractional order calculus

The fractional order calculus is an extension of the conventional integer order calculus. Instead of the first, second and, generally,  $n^{th}$  derivatives the order of derivative can be non-negative real number. Compared to the integer order differentiation, fractional order differentiation has no

straightforward graphical interpretation Podlubny (1999). The easiest way to describe the concept is to use the Laplace transform. The Laplace pair of an integer order differentiation of a function  $f(t)$  with zero initial conditions reads

$$\frac{d^p f(t)}{dt^p} \xleftrightarrow{\mathcal{L}} s^p F(s), \quad p \in \mathbb{N}, \quad s \in \mathbb{C}. \quad (1)$$

Nothing changes in (2) if instead of an integer  $p$  we take  $p \in \mathbb{R}^+$ . Unfortunately, the time domain equivalent of fractional differentiation is not so elegant and is represented by the Riemann-Liouville integral Podlubny (1999):

$$D_t^p f(t) = \frac{1}{\Gamma(n-p)} \frac{d^n}{dt^n} \left( \int_{-\infty}^t \frac{f(\tau)}{(t-\tau)^{p-n+1}} d\tau \right), \quad (2)$$

where  $(n-1) \leq p < n$ ,  $p$  determines the fractional differentiation order as in (1) and  $\Gamma(p)$  is the Gamma function.

### 2.2 Numerical evaluation

For many functions  $f(t)$ , the solution of the Riemann-Liouville integral (2) either does not exist in a closed form or includes the so-called Mittag-Leffler function. Moreover, if fractional derivatives of real signals need to be evaluated the integral (2) is needed. The common way of numerical approximation of the Riemann-Liouville (RL) integral is through the Grünwald-Letnikov (GL) scheme Podlubny (1999); Monje et al. (2010); Schmidt and Gaul (2000):

$$D_t^p f(t) = \lim_{N \rightarrow \infty} \left[ \left( \frac{t}{N} \right)^p \sum_{j=0}^{N-1} A_{j+1} f\left(t - j \frac{t}{N}\right) \right] \quad (3)$$

$$A_{j+1} = \frac{\Gamma(j-p)}{\Gamma(-p)\Gamma(j+1)} = \frac{j-1-p}{j} A_j.$$

Unfortunately, direct application of the GL numerical scheme (3) for fractional differentiation suffers the same drawback as ordinary numerical differentiation of noisy signals. At poor signal-to-noise ratios, the GL scheme provides numerically unstable results, thus limiting its usefulness for practical applications.

### 2.3 Problem definition

The problem of FOS identification is almost identical to that of integer order systems, i.e. the goal is to estimate the unknown  $a_i, b_j, \alpha_i, \beta_j$  of the following FOS:

$$y(t) + \sum_{i=1}^{N_a} a_i D_t^{\alpha_i} y(t) = \sum_{j=0}^{N_b} b_j D_t^{\beta_j} u(t), \quad N_b \leq N_a, \quad (4)$$

given the realisation of discrete values  $(u_i, y_i, i = 1, \dots, N)$  where  $\alpha_i \in \mathbb{R}^+$  and  $\beta_j \in \mathbb{R}^+$ . It should be noted that for a general case the powers  $\alpha_i$  and  $\beta_j$  are incommensurate<sup>1</sup>. By allowing  $\alpha_i \in \mathbb{N}$  and  $\beta_j \in \mathbb{N}$  the linear system (4) reduces to the integer order linear system.

The greatest issue of time domain FOS identification is numerical fractional order derivation of noisy signals thus precluding the applicability of GL numerical scheme. One possible remedy is by using the concept of modulating functions.

<sup>1</sup> Commensurate FOS is a special case and its identification represents somewhat simpler problem Malti et al. (2007).

### 3. MODULATING FUNCTIONS

*Definition 1.* (Preisig and Rippin (1993)). A MF  $\phi(t)$  of order  $n$  is a function defined on the interval  $[0, T]$  that satisfies the following properties:

- (1)  $\phi(t) \in \mathbb{C}^n([0, T])$
- (2)  $\phi^{(i)}(0) = \phi^{(i)}(T) = 0$  for  $i = 0, 1, \dots, n - 1$
- (3)  $D_t^\alpha \phi(t)$  exists,  $\forall 0 \leq \alpha \leq n$  and
- (4)  $D_t^\alpha \phi(t)|_{t=0} = 0, \forall 0 \leq \alpha \leq n$ .

A MF should have vanishing derivatives up to order  $n = \max\{\max(\alpha_i), \max(\beta_i)\}$ . Having a MF  $\phi(t)$  of order  $n \in \mathbb{N}$  and a function  $f(t)$  whose fractional derivative of order  $\alpha$  exists then the following relation involving convolution holds Preisig and Rippin (1993); Lorenzo and Hartley (2008):

$$\int_0^t D_t^\alpha f(\tau)\phi(t-\tau) d\tau = \int_0^t f(t-\tau)D_t^\alpha \phi(\tau) d\tau + \frac{c}{\Gamma(1-\alpha)} \int_0^T \phi(t-\tau)(\tau+a)^{-\alpha} d\tau - cD_t^{\alpha-1}\phi(t), \quad (5)$$

where  $f(t) = c$  for  $t \in [-a, 0], a > 0$ .

For initial conditions

$$\begin{aligned} D_t^{\alpha_i} y(t)|_{t=0} &= 0 \text{ for } i = 0, \dots, N_a \\ D_t^{\beta_j} u(t)|_{t=0} &= 0 \text{ for } j = 0, \dots, N_b, \end{aligned} \quad (6)$$

performing convolution with a MF  $\phi_m(t)$  on both sides using (5), the original problem (4) becomes:

$$\begin{aligned} \sum_{i=0}^{N_a} a_i \int_0^t f(t-\tau)D_t^{\alpha_i} \phi(\tau) d\tau \\ = \sum_{j=0}^{N_b} b_j \int_0^t u(t-\tau)D_t^{\beta_j} \phi(\tau) d\tau. \end{aligned} \quad (7)$$

The fractional derivatives in (7) include only MFs. Consequently, relation (5) transforms the original system in a form where fractional derivatives include noise free and more importantly known functions. Furthermore, for carefully selected MFs, the fractional derivatives exist in a closed form. The transformed system has the same set of parameters, however in the transformed form, it depends on the so-called modulated versions  $\tilde{u}(t)$  and  $\tilde{y}(t)$  of the original signals  $u(t)$  and  $y(t)$ .

There are many functions that satisfy the properties defined in Definition 1. One of the most commonly used is the so-called Cahen and Loeb function:

$$\phi_m(t) = \begin{cases} t^{n+m}(T-t)^n F(t) & \text{for } t \in [0, T] \\ 0 & \text{elsewhere,} \end{cases} \quad (8)$$

where  $F(t)$  is  $n - 1$  times differentiable. For the MF (8) the solution of the RL integral (2) exists in closed form Belkhatir and Laleg-Kirati (2017):

$$D_t^\alpha \phi(t) = \sum_{k=1}^m c_k \frac{\Gamma(K_k)}{\Gamma(K_k - \alpha)} t^{K_k - \alpha + 1}, \quad (9)$$

where  $K_k = M + \bar{a} - m + k + 2$ . Although the solution of (9) exists in closed form in suffers from the so-called ‘catastrophic cancellation’ (i.e. loss of significance). One way of avoiding this issue is by increasing the number of significant digits during the evaluation of (9). The cost is a substantial increase of the calculation time.

Conversely, the Puchov and Chinayev function offers a numerically efficient implementation even for substantially long signals. This function has the form of the first-order system with dead time as Preisig and Rippin (1993):

$$\phi(t) = \frac{a^4}{6}(t-T)^3 e^{-a(t-T)}, \quad (10)$$

where the parameter  $a$  determines the cut-off frequency of the filter, and  $T$  is sufficiently small. The Fourier transform of (10) exists in closed form:

$$H(j\omega) = \frac{a^4 (T^3 (a + i\omega)^3 - 3T^2 (a + i\omega)^2 + 6T (a + i\omega) - 6)}{(a + i\omega)^4 (T^3 a^3 - 3T^2 a^2 + 6T a - 6)} \quad (11)$$

### 4. THE PROPOSED PARAMETER ESTIMATION APPROACH

Recall that in the case for ordinary linear systems, where  $\alpha_i$  and the  $\beta_j$  are known integers, the estimation of model parameters can be done by one of numerous identification methods, see Garnier and Wang (2008). However, in fractional systems, neither fractional orders  $\alpha_i$  and  $\beta_j$ , nor its corresponding parameters are known. This section outlines the model parameter estimation, which includes (i.) estimation of the parameters  $a_i$  and  $b_j$ , and (ii.) estimation of corresponding fractional order.

In the context of SOFC’s and electrochemical energy systems in general, the system output  $u(t)$  corresponds to the measured voltage and the system’s input  $i(t)$  is the electrical current, therefore we rewrite (4) in the following form:

$$u(t) = \sum_{j=1}^{N_b} b_j D_t^{\beta_j} i(t) - \sum_{i=1}^{N_a} a_i D_t^{\alpha_i} u(t), N_a \geq N_b. \quad (12)$$

#### 4.1 Estimation of model parameters $a_i$ and $b_j$

Provided the fractional orders  $\alpha_i$  and  $\beta_j$  are known, one can evaluate fractional derivatives of the input/output signals employing e.g. (3). However, as already discussed in Section 3, this approach becomes inappropriate due to the issues related with numerical differentiation.

To overcome the issues related to the numerical differentiation, equation (12) is transformed into Fourier space and multiplied by function  $\Phi(j\omega)$  on the both sides to obtain

$$\begin{aligned} U(j\omega)\Phi(j\omega) &= \sum_{j=1}^{N_b} b_j I(j\omega)(j\omega)^{\beta_j} \Phi(j\omega) \\ &- \sum_{i=1}^{N_a} a_i U(j\omega)(j\omega)^{\alpha_i} \Phi(j\omega). \end{aligned} \quad (13)$$

There are no restrictions imposed on the selection of function  $\Phi(j\omega)$ . In this work the function of Puchov and Chinayev (11) is employed.

By reverting back to time domain we finally arrive to

$$u(t)*\phi(t) = \sum_{j=1}^{N_b} b_j i(t)*D_t^{\beta_j} \phi(t) - \sum_{i=1}^{N_a} a_i u(t)*D_t^{\alpha_i} \phi(t) \quad (14)$$

where  $*$  denotes the convolution operation.

For data collected every sampling period  $\Delta t$ , i.e.  $t \in \{0, \Delta t, 2\Delta t, \dots, N\Delta t\}$ , the above equation (14) can be written in matrix form

$$\mathbf{U} = \mathbf{A}_{\alpha,\beta} \mathbf{p}^T, \quad (15)$$

where  $\mathbf{U} = u(t) * \phi(t)$  is vector of length  $N$ ,  $\mathbf{p} = [b_{N_b}, \dots, b_1, a_{N_a}, \dots, a_1]$  is vector of length  $N_a + N_b$ , and matrix  $\mathbf{A}_{\alpha,\beta}$  of size  $N \times (N_a + N_b)$  contains the fractional derivatives of measured signals:

$$\mathbf{A}_{\alpha,\beta} = [\{i(t) * \mathcal{D}^{\beta_j} \phi(t)\} \{-u(t) * \mathcal{D}^{\alpha_i} \phi(t)\}]. \quad (16)$$

The unknown parameters in  $\mathbf{p}$  can be estimated from measured signals through linear least squares approach as:

$$\hat{\mathbf{p}} = (\mathbf{A}_{\alpha,\beta}^T \mathbf{A}_{\alpha,\beta})^{-1} \mathbf{A}_{\alpha,\beta}^T \mathbf{U}. \quad (17)$$

#### 4.2 Estimation of fractional orders

Provided the fractional orders  $\alpha_i$  and  $\beta_j$  are available and known, the model parameters in  $\mathbf{p}$  are easily evaluated by employing (17) following the procedure described above. Since that is not the case, an optimisation problem minimising the prediction error should be formulated.

The measurement matrix  $\mathbf{A}_{\alpha,\beta}$  in (15) is parameterised by fractional orders  $\alpha$  and  $\beta$ . For a given set of  $\mathbf{A}_{\alpha,\beta}$  the prediction error reads

$$\mathbf{e}(\alpha, \beta) = \mathbf{A}_{\alpha,\beta} \mathbf{p} - \mathbf{U}. \quad (18)$$

In order to find the optimal set of fractional orders, one needs to solve the following non-linear mathematical program:

$$\min_{\alpha, \beta} c(\alpha, \beta) \quad (19)$$

where we define optimisation criterion as mean squared error

$$c(\alpha, \beta) = \mathbf{e}(\alpha, \beta)^T \mathbf{e}(\alpha, \beta) = \sum_{n=0}^N \|\mathbf{A}_{\alpha,\beta}^{(n)} \mathbf{p} - \mathbf{U}^{(n)}\|_2^2, \quad (20)$$

where  $n$  sums over each row of the matrices. With optimisation defined as (20), the gradient can be easily found to be

$$\begin{bmatrix} \frac{\partial c(\alpha, \beta)}{\partial \alpha_i} \\ \vdots \\ \frac{\partial c(\alpha, \beta)}{\partial \beta_j} \\ \vdots \end{bmatrix} = \begin{bmatrix} 2 \sum_n (\mathbf{A}_{\alpha,\beta}^{(n)} \mathbf{p} - \mathbf{U}^{(n)}) \frac{\partial \mathbf{A}_{\alpha,\beta}^{(n)}}{\partial \alpha_i} \\ \vdots \\ 2 \sum_n (\mathbf{A}_{\alpha,\beta}^{(n)} \mathbf{p} - \mathbf{U}^{(n)}) \frac{\partial \mathbf{A}_{\alpha,\beta}^{(n)}}{\partial \beta_j} \\ \vdots \end{bmatrix} \quad (21)$$

where (index  $n$  is omitted)

$$\frac{\partial \mathbf{A}_{\alpha,\beta}}{\partial \alpha_j} = u(t) * \frac{\partial \mathcal{D}^{\alpha_i} \phi(t)}{\partial \alpha_i}, \quad \frac{\partial \mathbf{A}_{\alpha,\beta}}{\partial \beta_j} = i(t) * \frac{\partial \mathcal{D}^{\beta_i} \phi(t)}{\partial \beta_i}. \quad (22)$$

### 5. CASE STUDY

The experimental examinations of SOFC were performed at the Institute of Thermal Engineering at Graz University of Technology. The test rig assembly used for the experimental investigations is shown in Fig. 1.

The SOFC under test was an anode-supported with an active surface of 81 cm<sup>2</sup>. Nickel current collector was used on the anode side, while platinum was used to contact the cathode side. The cell housing was positioned in a temperature-programmed furnace and the operating

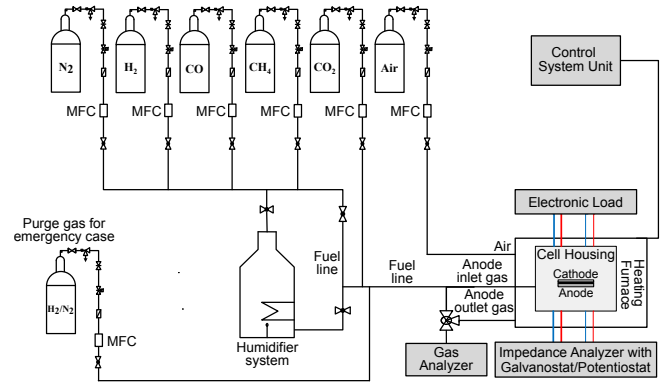


Fig. 1. SOFC test setup Subotić (2017)

temperature was held constant at 800°C. The selected current set point was 4 A.

The experiment went through two phases. In the first phase, the anode was fed with a dry H<sub>2</sub>/N<sub>2</sub> mixture, including 45 vol% H<sub>2</sub>, while the cathode was fuelled with air. The air and fuel flow in the co-flow conditions. The volume flow rate to the cell was set to 2.4 SLPM<sup>2</sup>, and 2 SLPM, for anode and cathode respectively. In the second phase the cell was intentionally poisoned while performing a run-to-failure experiment that lasted for 48 hours. For the purpose of the accelerated degradation, methane with S/C=0.5 was used as a fuel, thus forcing carbon deposition degradation phenomenon.

The system excitation consisted of two pseudo-random binary sequence (PRBS) signals PRBS1 and PRBS2 with cut-off frequencies of 1 Hz and 100 Hz respectively. The goal was to properly excite both slow and fast dynamic modes. The sampling frequency was  $f_s = 100$  kHz although the effective bandwidth was 10.8 kHz.

#### 5.1 Parameter estimation at the healthy state

The equivalent circuit model consists of 2 RQ elements. The rationale is twofold. When fuelled with pure hydrogen the EIS spectrum exhibits 3 semi arcs Bertei et al. (2016). Due to the limited bandwidth of our experimental equipment only the low and the middle frequency arcs are pronounced. In this particular case the model can be written as:

$$G_{FC}(s) = \frac{A}{\tau_a s^\alpha + 1} + \frac{B}{\tau_b s^\beta + 1} + R, \quad (23)$$

and the model parameters to be estimated include time constants  $\tau_a, \tau_b$ , static gains of the RQ elements  $A$  and  $B$ , serial resistance  $R$ , and finally the fractional orders  $\alpha$  and  $\beta$ .

Expanding the equation leads to the transfer function

$$G_{FC}(s) = \frac{b_3 s^{\alpha+\beta} + b_2 s^\beta + b_1 s^\alpha + b_0}{a_3 s^{\alpha+\beta} + a_2 s^\alpha + a_1 s^\beta + 1} \quad (24)$$

where the relations among parameters  $a_i, b_i$  and parameters of RQ elements  $R, A, \tau_a, B, \tau_b$  follow:

<sup>2</sup> SLPM stands for the *standard liters per minute*

$$\begin{aligned}
 b_3 &= R\tau_a\tau_b = p_1 & b_2 &= \tau_a(B + R) = p_2 \\
 b_1 &= \tau_b(A + R) = p_3 & b_0 &= A + B + R = p_4 \\
 a_3 &= \tau_a\tau_b = p_5 & a_2 &= \tau_a = p_6 \\
 a_1 &= \tau_b = p_7
 \end{aligned} \tag{25}$$

Following the parameter estimation algorithm described in 4 with constraints (25), the identified model reads

$$G_{FC} = \frac{1.18 \times 10^{-6}s^{1.72} + 0.0002s + 0.0001s^{0.72} + 0.02}{0.00029s^{1.72} + 0.07s^{0.72} + 0.004s + 1} \tag{26}$$

### 5.2 Validation of the SOFC model

The time domain identification results are shown in Fig. 2. During the first 3s the system was excited with the sum of PRBS1 and PRBS2, while in the time interval 3s – 12s only PRBS1 continued to excite the cell. From the results it is clearly visible that the identified system matches the measured signals very accurately.

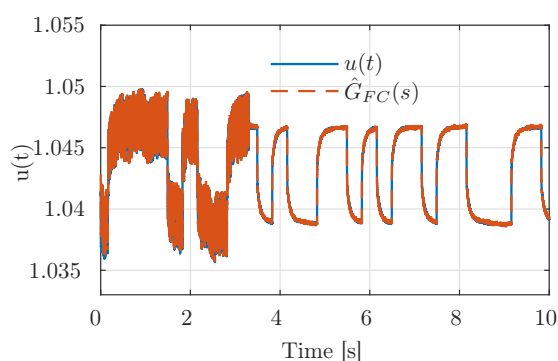


Fig. 2. The time domain identification results

Additional validation of the FOS model was done by comparing the Nyquist curves obtained from the estimated model parameters and the Nyquist curve obtained using the conventional sinusoidal perturbation. A set of 24 consecutive sinusoidal excitation signals (of frequencies from 0.1Hz-9kHz), with the duration of 7 periods each were applied. The complete duration of the entire excitation session was 133.33 seconds.

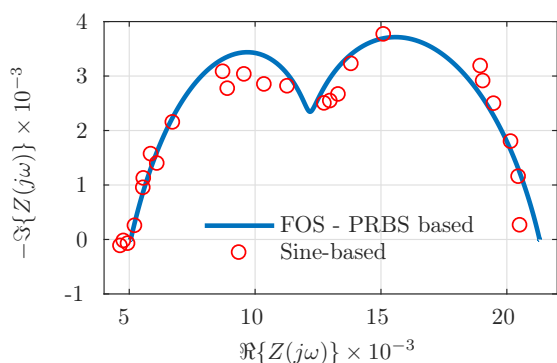


Fig. 3. Validation through comparison of the Nyquist curves

It is important to stress the following comments:

- (1) Substantially different perturbation times (133 s for repeated sinusoids versus 12 s of PRBS) result in almost the same Nyquist plots. Reductions of an order of magnitude are rather typical.

- (2) Short PRBS perturbation for FOS identification bears an additional advantage. Namely, by extending the experimental run the chance that external disturbances may affect the experiment increases. For example, small variations in the temperature, pressure, gas composition can result in fluctuations in the evaluated points on the Nyquist curve, which is clearly visible in Fig. 3.
- (3) Finally, unlike the classical sine-based EIS where the Nyquist curve is measured at discrete points, estimated FOS parameters provides fractional-order transfer function in a closed form as shown in Fig. 3.

The comparison in the frequency domain is an additional prove that the identified system is a clear match with the real one.

### 5.3 Application to condition monitoring of a single SOFC

Fig. 4 shows the estimated parameters of the fuel cell model for at six points during the run-to-failure experiment. The measurement indices denoted  $T_i$  are shown on the  $x$ -axis. The  $y$ -axis corresponds to particular parameter in (24). It is clearly seen that some of the parameters

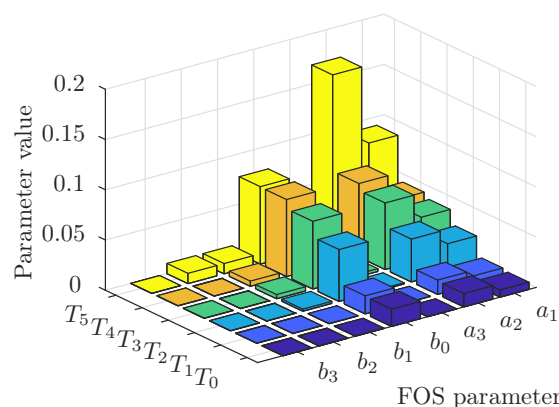


Fig. 4. Time evolution of the FOS parameters through short degradation experiment.

increase in value. This can be correlated with an increase of polarisation losses that, due to ongoing degradation inside the cell, increase significantly. Therefore, the estimated parameters can be directly employed for condition monitoring in the subsequent step.

## 6. CONCLUSION

A time domain approach to the estimation of incommensurate fractional order models is presented and its potential for health monitoring of SOFCs is demonstrated. Short persistent excitation by means of PRBS and pre-filtering of input and output signals by a modulating function allow for accurate identification of the linearized fuel cell dynamics. The approach is shown to outperform the conventional EIS approach in many aspects of which the substantially lower process excitation is the most striking. This makes the approach a viable solution for online health monitoring not only SOFCs but also other electrochemical conversion systems. In the work to follow a more systematic, data-driven, structural identification procedure able to select an optimal model from a set of the candidate models will

be investigated. No such results seem to be available at the time being.

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#### REFERENCES

- Barelli, L., Barluzzi, E., and Bidini, G. (2013). Diagnosis methodology and technique for solid oxide fuel cells: A review. *International Journal of Hydrogen Energy*, 38(12), 5060 – 5074.
- Belkhatir, Z. and Laleg-Kirati, T.M. (2017). Parameters and fractional differentiation orders estimation for linear continuous-time non-commensurate fractional order systems.
- Bertei, A., Ruiz-Trejo, E., Tariq, F., Yufit, V., Atkinson, A., and Brandon, N. (2016). Validation of a physically-based solid oxide fuel cell anode model combining 3d tomography and impedance spectroscopy. *International Journal of Hydrogen Energy*, 41(47), 22381 – 22393.
- Boškoski, P., Debenjak, A., and Mileva Boshkoska, B. (2017). *Fast Electrochemical Impedance Spectroscopy As a Statistical Condition Monitoring Tool*. SpringerBriefs in Applied Sciences and Technology. Springer. doi: 10.1007/978-3-319-53390-2.
- Dai, Y., Wei, Y., Hu, Y., and Wang, Y. (2016). Modulating function-based identification for fractional order systems. *Neurocomputing*, 173, 1959–1966.
- Eckert, M. (2017). *Modellbasierte Identifikation fraktionaler Systeme und ihre Anwendung auf die Lithium-Ionen-Zelle*. Ph.D. thesis, KIT-Fakultät für Elektrotechnik und Informationstechnik.
- Fuoss, R.M. and Kirkwood, J.G. (1941). Electrical properties of solids. viii. dipole moments in polyvinyl chloride-diphenyl systems\*. *Journal of the American Chemical Society*, 63(2), 385–394.
- Garnier, H. and Wang, L. (eds.) (2008). *Identification of Continuous-time Models from Sampled Data*. Advances in Industrial Control. Springer, London.
- Gehring, N. and Rudolph, J. (2017). An algebraic approach to identification of linear systems with fractional derivatives. In *IFAC World Congress*, 6388–6393.
- Heinzmann, M., Weber, A., and Ivers-Tiffée, E. (2018). Advanced impedance study of polymer electrolyte membrane single cells by means of distribution of relaxation times. *Journal of Power Sources*, 402, 24 – 33.
- Irvine, J.T. and Connor, P. (2013). *Solid Oxide Fuels Cells: Facts and Figures*. Springer-Verlag, London.
- Liu, B., Muroyama, H., Matsui, T., Tomida, K., Kabata, T., and Eguchi, K. (2010). Analysis of impedance spectra for segmented-in-series tubular solid oxide fuel cells. *Journal of The Electrochemical Society*, 157(12), B1858–B1864.
- Liu, D.Y. and Laleg-Kirati, T.M. (2015). Robust fractional order differentiators using generalized modulating functions method. *Signal Processing*, 107, 395 – 406.
- Liu, D.Y., Laleg-Kirati, T.M., Gibaru, O., and Perruquetti, W. (2013). Identification of fractional order systems using modulating functions method. In *American Control Conference (ACC), 2013*, 1679–1684. IEEE.
- Lorenzo, C.F. and Hartley, T.T. (2008). Initialization of fractional-order operators and fractional differential equations. *Journal of Computational and Nonlinear Dynamics*, 3(2), 021101–021101–9.
- Malti, R., Victor, S., Nicolas, O., and Oustaloup, A. (2007). System identification using fractional models: State of the art. In *ASME International Design Engineering Technical Conferences*.
- Marra, D., Pianese, C., Polverino, P., and Sorrentino, M. (2016). *Models for Solid Oxide Fuel Cell Systems*. Springer-Verlag, London.
- Monje, C., Chen, Y., Vinagre, B., Xue, D., and Feliu-Batlle, V. (2010). *Fractional-order Systems and Controls*. Springer.
- Niya, S.M.R., Phillips, R.K., and Hoorfar, M. (2016). Process modeling of the impedance characteristics of proton exchange membrane fuel cells. *Electrochimica Acta*, 191, 594 – 605.
- Pahon, E., Steiner, N.Y., Jemei, S., Hissel, D., Péra, M., Wang, K., and Moçoteguy, P. (2016). Solid oxide fuel cell fault diagnosis and ageing estimation based on wavelet transform approach. *International Journal of Hydrogen Energy*, 41(31), 13678 – 13687.
- Podlubny, I. (1999). *Fractional Differential Equations*, volume 198 of *Mathematics in Science and Engineering*. Elsevier.
- Polverino, P., Sorrentino, M., and Pianese, C. (2017). A model-based diagnostic technique to enhance faults isolability in solid oxide fuel cell systems. *Applied Energy*, 204, 1198 – 1214.
- Preisig, H.A. and Rippin, D.W.T. (1993). Theory and application of the modulating function method—i. review and theory of the method and theory of the spline-type modulating functions. *Computers & Chemical Engineering*, 17(1), 1–16.
- Sabatier, J., Lanusse, P., Melchior, P., and Oustaloup, A. (2015). *Fractional Order Differentiation and Robust Control Design*. Springer Science.
- Schmidt, A. and Gaul, L. (2000). Application of fractional calculus to viscoelastically damped structures in the finite element method. In *International Conference on Structural Dynamics Modelling*, 297–306.
- Sorce, A., Greco, A., Magistri, L., and Costamagna, P. (2014). Fdi oriented modeling of an experimental soft system, model validation and simulation of faulty states. *Applied Energy*, 136, 894 – 908.
- Sorrentino, M., Marra, D., Pianese, C., Guida, M., Postiglione, F., Wang, K., and Pohjoranta, A. (2014). On the use of neural networks and statistical tools for nonlinear modeling and on-field diagnosis of solid oxide fuel cell stacks. *Energy Procedia*, 45, 298 – 307. ATI 2013 - 68th Conference of the Italian Thermal Machines Engineering Association.
- Subotić, V. (2017). *Detection of carbon depositions and development of novel regeneration approaches for solid oxide fuel cells*. Ph.D. thesis, TU Graz, Austria.