

# An Online Reliable Assessment System

Dani Juricic, Gjorgji Nusev, Bostjan Dolenc, Pavle Boskoski  
Jozef Stefan Institute  
Department of Systems and Control  
Jamova 39, 1000 Ljubljana  
Slovenia



**ABSTRACT:** *For an extensive commercial application the reliability of fuel cells has to be improved. For a reliable online assessment, the accurate parameters of the internal conditions have to be checked. Besides, we further suggest the capturing the full information on the estimates i.e. their probability density functions. In this work, we have presented the assorted results of the experiments conducted on a short-stack solid oxide fuel cell system.*

**Keywords:** Solid Oxide Fuel Cell, Bayesian Estimation, Fault Detection, Equivalent Circuit Model

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## 1. Introduction

Solid-oxide fuel cell systems (SOFCs) are devices that perform the conversion of chemical energy of fuels into electrical energy and heat. SOFCs can operate on a broad power range from a few kW up to several hundred kW or even MW, thus covering the needs of the residential and other stationary applications. The SOFC technology exhibits over 50% electrical efficiency (in less than 60% of cases), which is an advantage for integration into low-energy buildings. It is a trend for today's and tomorrow's construction where heat demand will decrease and the need for electricity increased.

Although promising results have been achieved in the SOFC, the main problem for their commercial use remain its operational reliability and unsatisfactory life expectancy. Due to the high operating temperature (700-800 ° C), the systems are more complex and the problem is the installation of sensors to monitor the situation inside. As far as life expectancy is concerned, the Julich Development Center in December 2015 achieved a world record of malt, which lasted 70,000 hours at an average degradation rate of 0.6% / 1000 hours of operation. The test was performed under laboratory conditions with a highly instrumented short fund (SS), while long-term tests on real systems on the field do not yet exist.

A way to make SOFC technology more competitive on the market is to use techniques for early detection of injuries during the operation. Damage should be detected as soon as possible so that corrective measures can be taken in good time. The conventional approaches are electrochemical impedance spectra, relaxation time distribution (DRT), and evaluation of the

equivalent circuit of the replacement model parameters (ECM). A change in the internal state of the cells, whether it is a degradation mechanism or a fault, affects the impedance curves and the associated parameters of the ECM circuits. Since SOFCs are characterized by eigenmodes on a broad range, meaningful characterization requires excitation from *mHz* to tens of *kHz*. Classically, the system is successively probed with a sinusoidal current of the selected frequency and a small amplitude around the operating point in order to evaluate the impedance (Nyquist) curve from the amplitude and phase of current and the voltage. The problem is that low frequencies require a long time to perform. That significantly prolong the test, especially if a number of experiments have to be repeated at low frequencies. An additional problem is that during the long tests, the potential of the disturbances to spoil the results increases.

Therefore, we proposed an approach that uses broadband excitation signals, i.e. pseudo-random binary noise (PRBS). From the complex wavelength analysis (CWT) of the input and output signals, it is possible to calculate the impedance characteristics of the system with resolution defined by the sampling rate. From impedance, the parameters of the equivalent circuit (ECM) are evaluated. Because it is about optimizing criterion functions that are poorly-conditioned, the evaluation process is done in two steps, first by using the genetic algorithm the most promising solution is found, and then using the simplex method to find the optimal in its vicinity. To find the distribution of the estimated parameters, the Markov Chain Monte Carlo (MCMC) approach is used.

The process was evaluated on a short 6-cell line of solidoxide fuel cells (SOFC) for a period of 3600 hours. The first results obtained on a short stack are presented.

## 2. Preliminaries

### 2.1. Data Acquisition

To evaluate electrochemical impedance spectra (EIS) requires voltage and current data. In the context of electrochemical energy systems, the highest frequency at which EIS is analysed is usually in the interval of 10kHz. Typically, multi-channel data acquisition systems offer interlaced sampling, which inevitably introduces error in the phase estimation. errors induced by the interlacing approach.

In order to guarantee a viable fit, the first step is to validate the correctness of the impedance data. For linear, causal and stable systems the impedance curve is a complex analytic function in the upper half-plane. For such functions, the real and imaginary part are linked through the so-called Kramers-Kronig (KK) relations [2]. Later Bode [3] was the first to successfully apply these relations to electrical impedances and since then it has become a basic tool for checking the validity of the obtained data.

The *KK* relations linking the real and imaginary components are:

$$\Im(Z(\omega)) = -\frac{2\omega}{\pi} \int_0^{\infty} \frac{\Re(Z(x)) - \Re(Z(\omega))}{x^2 - \omega^2} dx \quad (1)$$

The relations (1) requires integration up to  $\infty$ , which for finite length discrete measurements imposes inevitable error. A solution that circumvents this numerical calculation issue is provided by the so-called *Z-HIT* method [4]. The method allows calculation of the amplitude spectrum  $H(\omega)$  from the phase spectrum  $\phi(\omega)$  as:

$$\log |H(\omega_0)| \approx \text{const} + \frac{2}{\pi} \int_{\omega_{max}}^{\omega_0} \phi(\omega) d \log \omega + \gamma \frac{d\phi(\omega_0)}{d \log \omega} \quad (2)$$

where  $\gamma = -\pi/6$  and  $\omega_0 \in [\omega_{min}, \omega_{max}]$ . Derivation  $d\phi(\omega_0)/d \log \omega$  was calculated using the Savitzky–Golay filter. The integration constant is determined by the least squares fit. An example of the performance of the ZHIT test is shown in Figure 1.

### 2.2. Linearized SOFC Dynamic Model

The model of the linearized SOFC system dynamics in *complex space* can be expressed by the transfer function

$$Z(j\omega) = R_s + \sum_{i=1}^{N_D} \frac{R_i}{\tau_i(j\omega)^{\alpha_i + 1}} \quad (3)$$

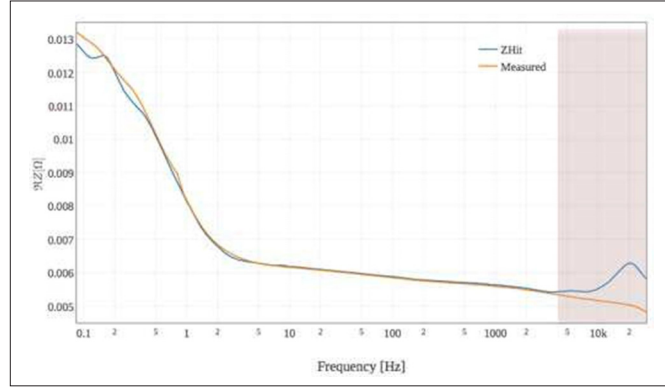


Figure 1. Comparison between measured modulus and modulus reconstructed from phase by using the ZHIT rule. The ZHIT test starts showing inconsistencies after cca. 5kHz

where  $0 < \alpha \leq 1$  stands for fractional derivative. Starting from measured input/output data, obtained during probing with the PRBS, the characterisation of the SOFC is performed by estimating the parameters of the model (3). This can be done either in the time domain by using the approach presented in [5] or in the frequency domain. The latter option relies on the evaluation of the transfer function (3) after evaluating the complex wavelet transform of voltage and current [1]. For the sake of simplicity, in the sequel, we will stick to the second option.

To capture the full information on the parameters it is necessary to set up the estimation problem in the probabilistic framework. By doing so, not only point estimates of the model parameters are obtained, but also their corresponding uncertainties. Uncertainties are much too often neglected in practice, however, they indirectly bear a valuable piece of information as a resultant of the quality of measurements, noise conditions, the importance of a parameter etc. For that purpose, the Bayesian approach based on MCMC approach is adopted.

### 3. Bayesian Estimation of the Fractional Order System Model

The idea of a Bayesian approach is to fuse *prior information* on an unknown variable  $\theta$  with the information on that variable contained in the data  $D = \{d_1, \dots, d_n\}$ .

Given a model and data  $D$ , the posterior distribution of the *unknown* parameters  $\theta$  can be evaluated via the Bayes rule:

$$p(\theta | D) = \frac{p(D|\theta) \cdot p(\theta)}{\int_{-\infty}^{\infty} p(D|\theta) p(\theta) d\theta} \quad (4)$$

where  $p(D|\theta)$  is a likelihood,  $p(\theta)$  is prior, and  $\int p(D|\theta) p(\theta) d\theta$  is called a marginal likelihood or model evidence.

Selecting the model structure in Bayesian inference is the most crucial part of the modelling procedure. Since the model structure of the fuel cell impedance is well defined in the frequency space  $\{\omega_1, \dots, \omega_N\}$ , one can easily construct the likelihood function:

$$p(D|\theta) = \prod_i p(\omega_i|\theta)$$

where we assume that measurement points  $i$  on the Nyquist curves are independent and that  $p(\omega_i|\theta)$  defined in the following way

$$p(\omega_i|\theta) = \frac{1}{\sigma \sqrt{2\pi}} \exp \left( -\frac{(y_i - \Re \{Z(j\omega_i|\theta)\})^2}{2\sigma^2} \right)$$

The parameter vector  $\theta$  contains the resistances  $R_k$ , time constants  $\tau_k$  and rational exponents  $a_k$  for each of the  $k^{\text{th}}$  element in the ECM. In the above equation,  $y_i$  denotes the real part of the measured point on the Nyquist curve at a frequency  $\omega_i$ , i.e.  $y_i =$

$\Re(Z(j\omega))$ .

Note that, in order to estimate the parameters  $\theta$  in (3), only one component of the complex impedance is required, hence  $\Re\{\cdot\}$ . Due to the Kramers-Kronig relation, it is enough to consider only the information either in real or imaginary parts of the complex value.

The prior probability of the parameter  $p(\theta)$  is generally used to incorporate any prior knowledge about the modelled system. For the purposes of condition monitoring, it can incorporate knowledge about the model parameters from measurements that were done at some previous time instance e.g. at commissioning of the system. In this work, without any prior knowledge, an un-informative truncated normal prior  $p(\theta)$  was used for the model parameters  $R_i$  and  $\tau_i$ , since these parameters always take positive values. On the other hand, uniform distribution was used as the prior parameters.

Having specified the prior and the likelihood of the data, the posterior distribution of the model parameters can be inferred by employing (4). However, with complex models, the analytic solution for  $p(\theta/D)$  is rarely possible due to the integral in the denominator. Readily, there are few options that resolve this issue by employing numerical methods. The integral can be solved by using grid approximation. However, this becomes computationally demanding with higher dimensions of the parameter space. On the other hand, the Markov Chain Monte Carlo (MCMC) simulations can be used to sample directly from the posterior distribution without the need to evaluate  $p(\theta/D)$ . In such a case, only the proportional part of the equation (3) is required to be numerically tractable:

$$p(\theta/D) \propto p(D|\theta) p(\theta).$$

More on MCMC methods and Bayesian inference can be found in [6]. The demonstration is available online to fully visualise the approach at the link <https://chi-feng.github.io/mcmc-demo/>

## 4. Experimental Results

### 4.1. The Experiment in Brief

A SOLIDpower SOFC short stack operating at 750°C was used in the experiment. The stack consisted of six planar anodesupported cells which were installed in an insulated ceramic housing. The active area of a single cell was 80 cm<sup>2</sup>. The SOFC short stack was fed with a mixture of hydrogen and nitrogen with a flow rate  $H_2/N_2=0.216/0.144$  NI h<sup>-2</sup>1cm<sup>-2</sup> whereas the air flow rate was 4 NI h<sup>-1</sup>1cm<sup>-2</sup>. Stack was operated at a nominal current of 32 A (0.4 A cm<sup>-2</sup>) with fuel utilization  $FU = 77.5\%$ . The experiment took  $\approx 3600$  hours. During this period, characterization was automatically performed on a regular basis by employing both conventional sinusoidal excitation and PRBS waveforms excitation every 6 hours. Hence a dataset with voltage and current recordings acquired at 600 measurement sessions is obtained.

The stack was first run at nominal conditions for 240 h (40 measurement sessions). After warming up, the first fuel starvation session was performed by gradually raising the fuel utilization (FU) stepwise each 24 h starting from 77.5% up to 92.5% (Figure 2 event E2). This was done by decreasing H2 flow rate while keeping the current density constant. The second fuel starvation protocol was performed also at the same FU steps as in the first situation, but by keeping H2 flow rate constant while increasing the current density (Figure 5 event E3). The current was increased according to the following sequence 32 A, 34.06 A, 36.15 A, 38.2 A for each of the FU.

### 4.2. Results Obtained on Data from One Measurement Session on a Single Cell

We will first demonstrate the application of Bayesian inference to the FOS parameter estimation on a single cell. Figure 3 shows the marginal posterior densities of model parameters. The first row shows three-time constants  $\tau_i$ , the second row shows resistances  $R_i$ , and the third row shows the fractional orders  $\alpha_i$ . Note that the last row displays only serial resistance. One can notice that the distributions of some of the parameters are far from normal. That is particularly the case for  $\alpha_1$ ,  $\alpha_2$ ,  $R_1$ , and  $R_2$ .

### 4.3. Time Evolution of the Estimated Parameters of Cell #3

We will now see how the parameters of the model of cell #3 evolve over time. The time index refers to the measurement

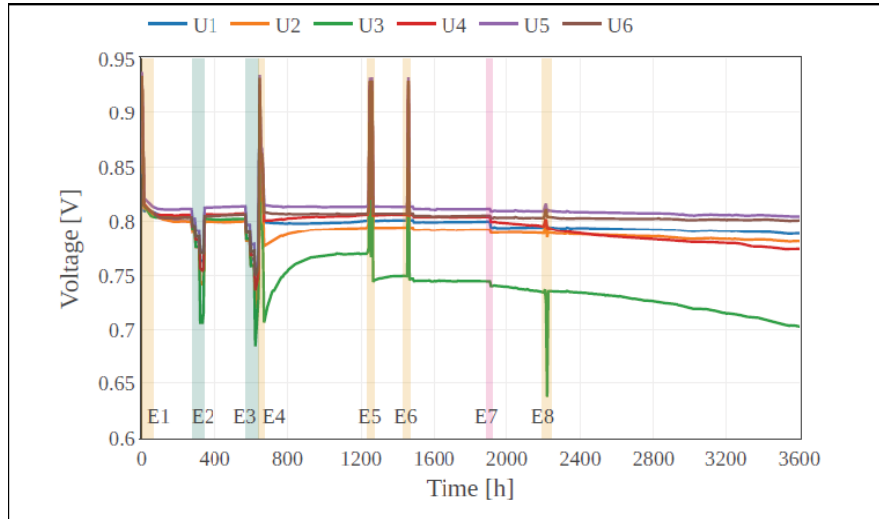


Figure 2. Cell voltages during the experiment

session. Yellow strips mark the intervals of too high fuel utilization.

The increase of fuel utilization (events E2) causes the resistances  $R_3$  (related to the slowest mode  $\tau_3 \sim 1.2s$ ) and  $R_s$  to blow up (Figure 4). That can be seen also as blow up in the lowfrequency part of the Nyquist curve (not shown here). The estimates take

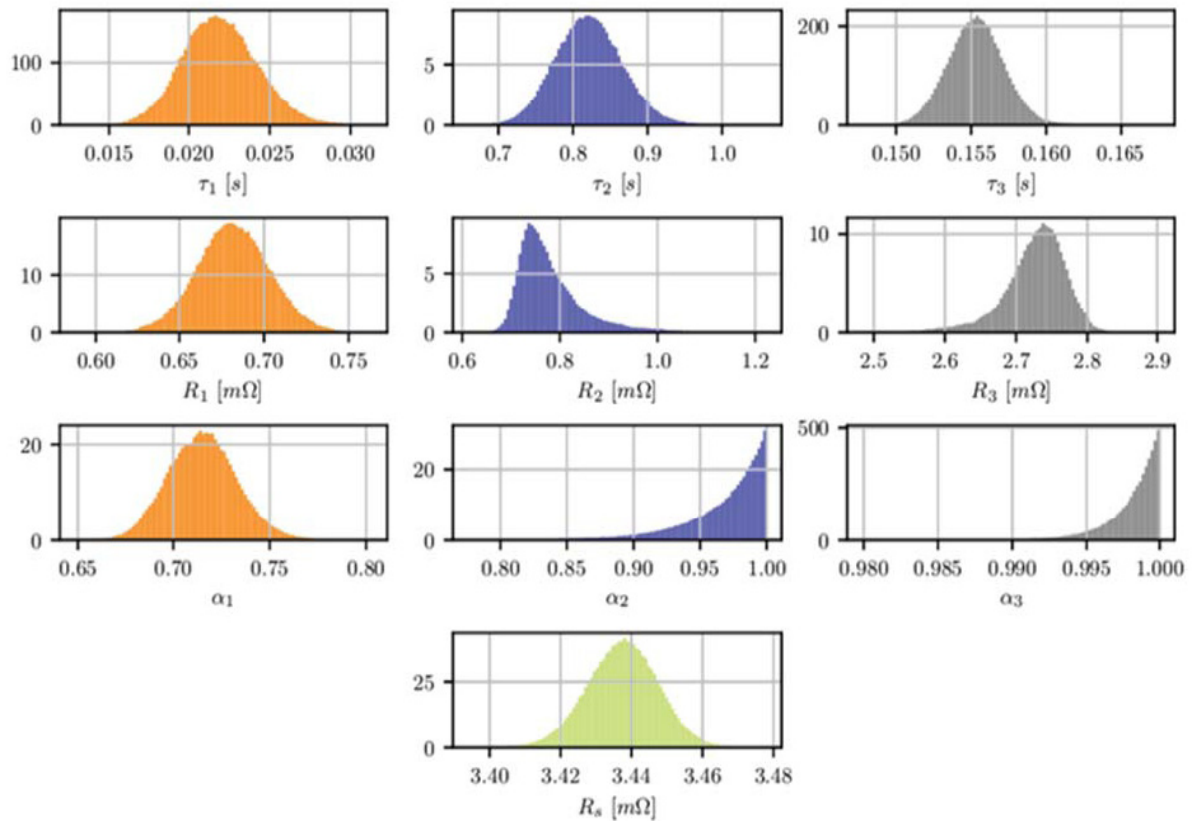


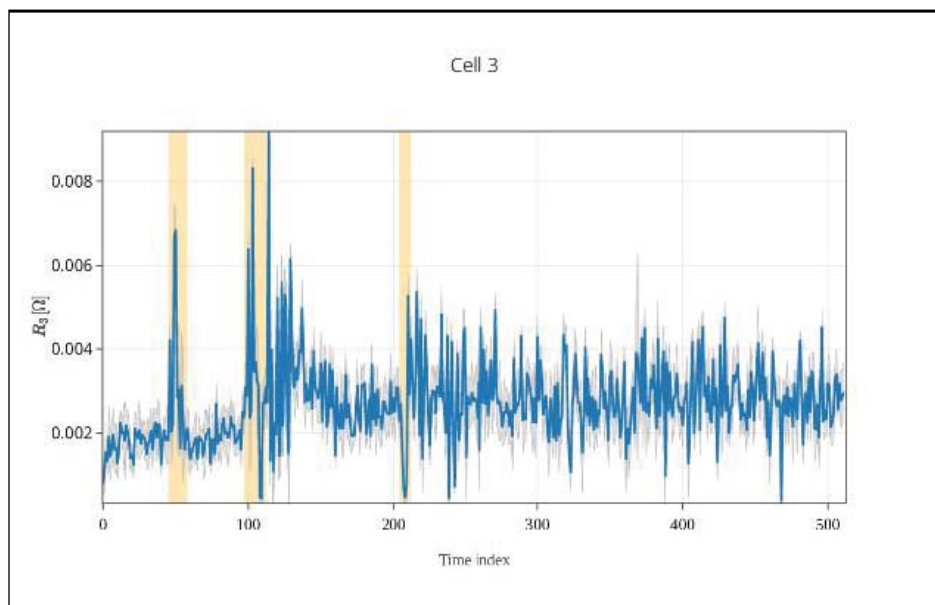
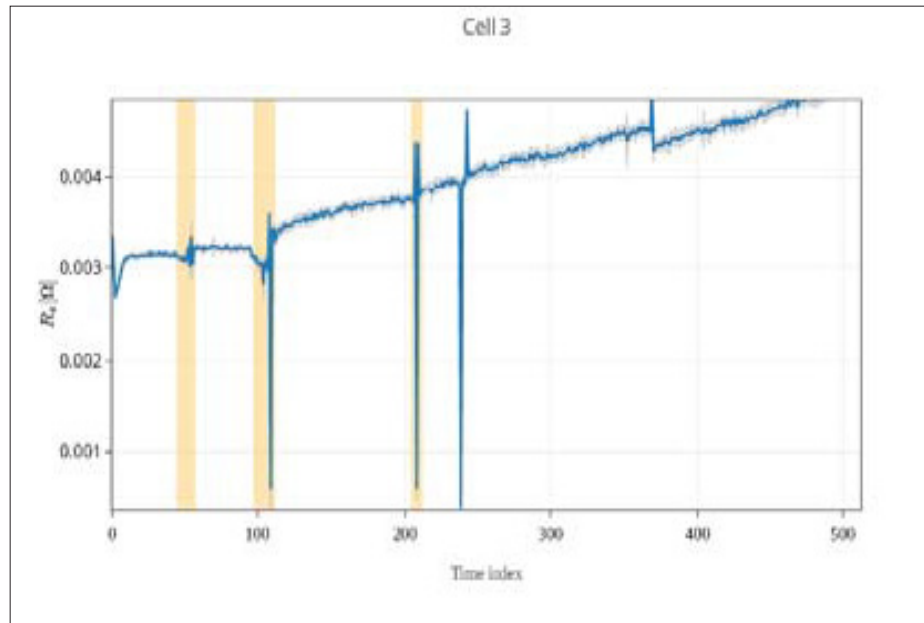
Figure 3. Results of the MCMC approach when applied to the data acquired during one measurement session on cell #3. The plots present the posterior distributions of model parameters from a model with 3 RQ elements

unusual values also in cases of incident events like hydrogen supply shutdown (HSS1, HSS2) and power supply shutdown (PSS). The model reacts also to the migration of the equipment into another laboratory.

The estimates of the parameters over time are not smooth but fluctuates (c.f. Figure 4). For instance, the estimates of  $R_s$  are relatively smooth while the estimates of  $R_3$  and  $\tau_3$  are rather “noisy”. Moreover, note that the uncertainty region of the estimates ( $\pm\sigma$ ) is relatively narrow, meaning that the parameter estimation algorithm ends up with rather highly reliable estimates. The explanation for such results should be sought in two limited quality of current sensor as well as fluctuations in fuel flows.

### 5. Conclusion

We presented a Bayesian approach to the parameter estimation of the linearized model of solid oxide fuel cell dynamics. From the change in the marginal pdf's from their reference forms, it will be possible to detect changes in the internal condition of the system.



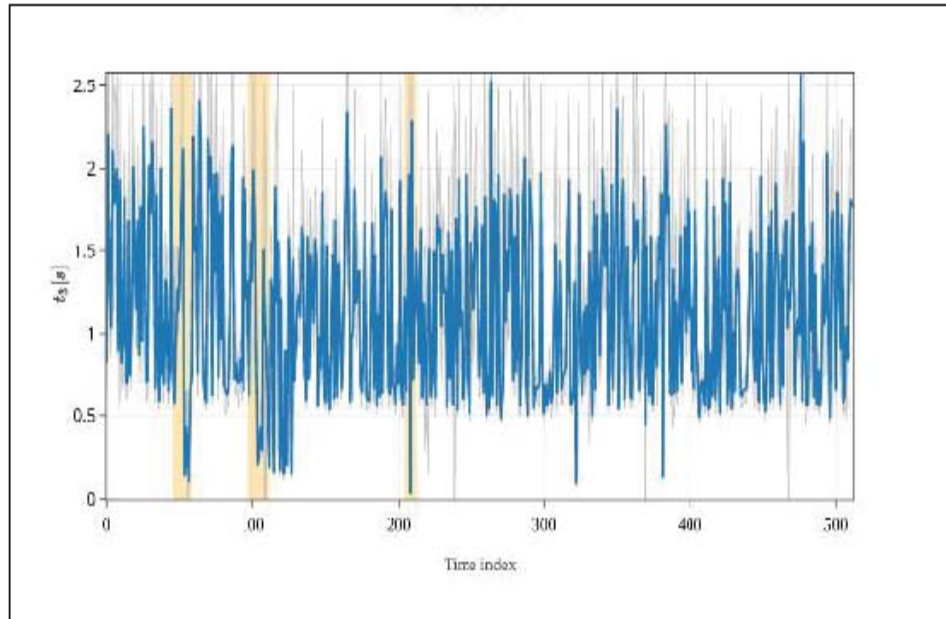


Figure 4. An excerpt from evolution of the model parameters with focus on  $R_s$  and  $R_3$  and  $T_3$

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