Frequencies selection for accelerated CNLS parameter identification of anticorrosion coatings

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Abstract- The paper presents modified CNLS impedance spectrum fitting method of parameter identification of technical objects. The number of test frequencies is reduced to the number of parameters and the test frequencies are set according to various criteria. As a test object, the model of anticorrosion coating has been chosen. The simulated identification results are discussed in terms of identification measurement time reduction and method accuracy.

I. Introduction

Modelling of technical objects (anticorrosion coatings [1], materials [2], sensors [3], reinforced concrete constructions [4]) and biological objects (skin and physiological fluids [5], tissues [6]) with electrical circuits (multi-element two-terminal networks) is currently becoming more and more popular. Such modelling allows simulating these objects, performance evaluation, monitoring and diagnosis of their state with well-developed tools and methods designed for electrical circuits.

In order to identify object impedance parameters, the impedance spectroscopy methods are used. They are based on impedance spectrum measurement in a wide frequency range. The measurement is usually carried point-by-point with a single frequency impedance analyser. The parameters are found by fitting the parameter dependant object model to the measured impedance spectrum [7], usually with Complex Non-Linear Least Square (CNLS) fitting algorithm [8].

Although very popular and almost being the standard in impedance parameter identification, the CNLS method has some serious drawbacks. Firstly, without using the knowledge about an expected object topology and parameters, the required range of impedance spectrum begins with low or very low frequencies (in case of anticorrosion coatings order of mHz or μHz), and the usual number of impedance spectrum points is about 3-5/decade. Secondly, the point-by-point impedance measurement method, for such number and range of spectrum points is very time consuming.

II. Objectives

As the long measurement time (order of hours) is inconvenient in field measurements (due to both technical and economical reasons) there is a strong need for acceleration of identification measurements of technical objects' impedance models [9]. To achieve that, both impedance spectrum measurements and identification methods have to be modified. The acceleration of impedance spectrum measurement, for a given set of frequencies via multisine stimulation and various analysis methods has been already discussed in [10-11]. In this paper, the modification of CNLS parameter identification method oriented for such measurement is being proposed.

Usually, the number of CNLS identified circuit's parameters is several times smaller than the number of impedance spectrum measurement points. That over-determination of identification equations is beneficious in case of scientific research – the measurement can be made before choosing an equivalent electric circuit and without any assumptions on object parameters. However, for diagnostic measurement of a well-known object degrading over time (like anticorrosion coating), with a given model and expected (assumed) values of parameters – that redundancy leads to unnecessary long measurement time.

In this paper, we propose and test, by means of simulation, a modification to conventional CNLS fitting method for diagnostic measurement of technical objects. The novelty lies in using only a limited number of test frequencies (equal to number of identified parameters). They are set according to criteria based on various approaches to the sensitivity analysis of model's impedance function. As a test object the anticorrosion coating equivalent circuit has been chosen. The identification results for different criteria and for conventional multi-frequency CNLS method are compared in terms of identification uncertainty and measurement time reduction.

III. Methodology

The new approach to CNLS fitting, with a limited number of impedance spectrum test frequencies, selected by developed criteria has been tested by means of simulation. The Beaunier's model of anticorrosion coating has been chosen, presented in. Fig.1.

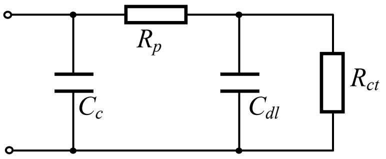

Figure 1. Test object - Beaunier's equivalent circuit of anticorrosion coating.

The impedance function for that circuit is:

$$Z(j\omega)=\frac{R_p+R_{ct}+j\omega\cdot C_{dl}\cdot R_p\cdot R_{ct}}{1-\omega^2\cdot C_{dl}\cdot C_C\cdot R_p\cdot R_{ct}+j\omega\cdot C_{dl}\cdot R_{ct}+j\omega\cdot C_C\cdot R_{ct}+j\omega\cdot C_C\cdot R_p}, \qquad (1)$$

dependant on vector of parameters $\boldsymbol{p}=[C_c\ R_p\ C_{dl}\ R_{ct}]^{\mathrm{T}}$.

Five combinations of model parameters have been set, corresponding to five degradation stages of anticorrosion coating, starting from nominal A, through under paint corrosion stages B-D, up to stage E, when the coating is delaminated, and penetrated by water. The parameters are presented in Tab.1.

Table 1. Parameters of anticorrosion coatings at different stages od degradation.

Object	R_p	R_{ct}	C_c	C_{dl}
Stage A	100GΩ	100GΩ	10pF	100pF
Stage B	10GΩ	10GΩ	100pF	1nF
Stage C	10GΩ	1GΩ	1nF	10nF
Stage D	1GΩ	0,1GΩ	1nF	100nF
Stage E	1GΩ	0,1GΩ	1nF	1μF

The impedance spectrum for the nominal (stage A) set of parameters is presented in Fig. 2.

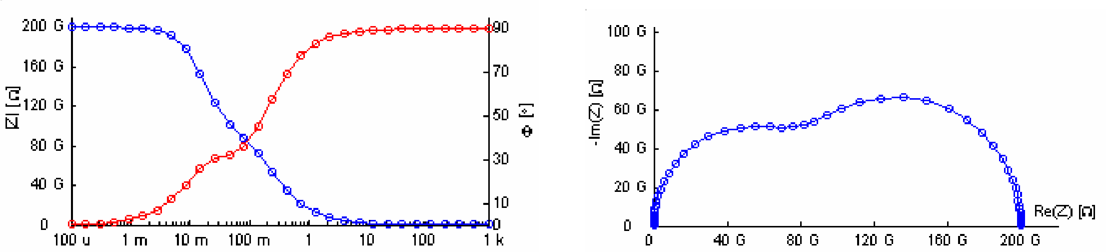

Figure 2. Bode and Nyquist plots of impedance spectrum of coating model in stage A.

In order to test the propagation of measurement uncertainty to the vector of parameters $\boldsymbol{p}$, the Monte Carlo approach has been used. The nominal impedance measurement values have been calculated from the impedance function with a given model (A-E) parameters. Then, the measurement uncertainty has been taken into consideration, by adding a random 1% multiplicative error of modulus and 0.001° random additive phase angle error, both drawn from rectangular distributions. Such an "uncertaintization" of a simulated result has been repeated 100 times, thus producing a set of 100 simulated impedance measurement data. For the set of data, the CNLS identification has been done, producing a series of identified parameters, followed by calculation of mean and standard deviation of relative identification error; speaking precisely: the relative identification error of mean and relative standard deviation. First value is a good indicator if the iterative identification process goes well. Second one is the approximant of identification uncertainty for a specified uncertainty of measurement.

The CNLS iterative method requires assuming start values – in this case, the nominal values of anticorrosion coating (stage A) have been chosen. Also, the test frequencies have been selected on the basis of nominal circuit impedance. That corresponds to a situation of a diagnosing a coating in an unknown state, whereas the previous test had been done for the nominal state of coating.

The simulated measurement data sets have been prepared in Matlab environment. The CNLS fitting has been done by a Macdonald LEVM program. In order to automate the simulation for more than 2000 fittings, the LEVM program was run in a batch mode, controlled by a dedicated application written in LabWindows/CVI. The dedicated software has prepared the input data files for batch mode of LEVM and also post-processed the LEVM output files. It has also calculated the statistical parameters of series of identification results.

IV. Description of Test Frequencies Selection Criteria

In order to choose test frequencies, the selection criteria have to be defined. The criteria were formed on the basis of the sensitivity of object impedance function to the identified parameters. In this case, the modulus of small-signal relative sensitivity [12] was taken into consideration. It was calculated with differential calculus, according to formula:

$$S_{p_k} = \left| \frac{\dfrac{\Delta Z}{Z}}{\dfrac{\Delta p_k}{p_k}} \right| = \left| \frac{\partial Z}{\partial p_k} \cdot \frac{p_k}{Z} \right| . \tag{2}$$

The plot of test object sensitivity to parameters of model against frequency is presented in Fig. 3.

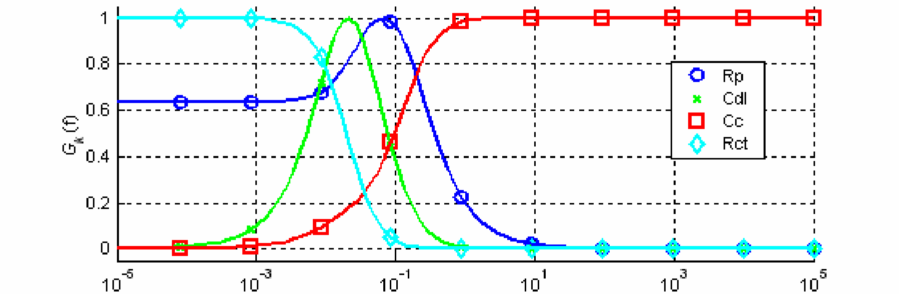

Figure 3. Small-signal sensitivity of model of coating in stage A.

The criteria proposed, allow to find the vector of test frequencies $\boldsymbol{f}=[f_1,f_2,f_3,f_4]^T$ of length equal to number of identified parameters, by assuming that each frequency is optimal for one parameter, so the vector $\boldsymbol{f}$ is exactly a $\boldsymbol{f}=[f_{Cc},f_{Rp},f_{Cdl},f_{Rct}]^T$. Each frequency in a vector can be determinated by finding a maximum of objective function $G_k(x)$, defined for each parameter, on a limited frequency range from f_{min} to f_{max}. The formal notation for the $G_k(x)$ being the curve with a maximum:

$$f_x = f_k \Leftrightarrow G_k(f_x) = \max_{f_{\min}<f<f_{\max}} \left[G_k(f)\right]. \tag{3}$$

If the $G_i(x)$ does not have an extremum (it is asymptotic), the optimal frequency is evaluated as a point where the objective function drops by a given ε, e.g. 1% :

$$f_x = f_k \Leftrightarrow G_k(f_x) = (1-\varepsilon) \max_{f_{\min}<f<f_{\max}} \left[G_k(f)\right]. \tag{4}$$

The first criterion used (noted C_1) was a maximum of impedance sensitivity for a given parameter:

$$G_k(f) = S_{p_k}(f). \tag{5}$$

In case of asymptotic curves, which have not had an extremum, the optimal frequency was assumed as the frequency for which the sensitivity drops 1% below an asymptote level:

$$f_x = f_k \Leftrightarrow S_{p_k}(f_x) = (1-\varepsilon) \lim_{f \to f_{\max}} \left(S_{p_k}\right) \vee S_{p_k}(f_x) = (1-\varepsilon) \lim_{f \to f_{\min}} \left(S_{p_k}\right). \tag{6}$$

Second criterion (C_2) was formulated to consider not only the maximum sensitivity for a given parameter, but also to minimize the influence of other model parameters.

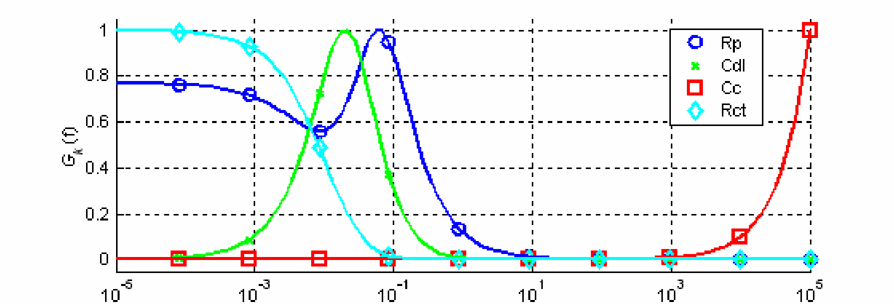

Figure 4. Plot of the objective function for criterion C_2.

Objective function was constructed by dividing the value of sensitivity to a parameter *k* by a sum of sensitivities to parameters other than *k*. For visualisation purposes, every curve $G_k(f)$ was normalized to its maximum:

$$G_k(f)=\left\|\frac{S_{p_k}(f)}{S_{p_1}(f)+\ldots+S_{p_{k-1}}(f)+S_{p_{k+1}}(f)+\ldots+S_{p_N}(f)}\right\|. \tag{7}$$

The normalized plot for criterion C_2 is presented in Fig. 4.

The components of denominator in (6) are not influencing the objective function equally, as the ranges of their values are different, what can be seen in Fig. 2.

Thus, the third criterion has been proposed (noted C_3), with the sensitivities in denominator being normalized. The objective function for criteria C_3 is:

$$G_k(f)=\left\|\frac{\left\|S_{p_k}(f)\right\|}{\left\|S_{p_1}(f)\right\|+\ldots+\left\|S_{p_{k-1}}(f)\right\|+\left\|S_{p_{k+1}}(f)\right\|+\ldots+\left\|S_{p_N}(f)\right\|}\right\|, \tag{8}$$

and its plot is presented in Fig. 5.

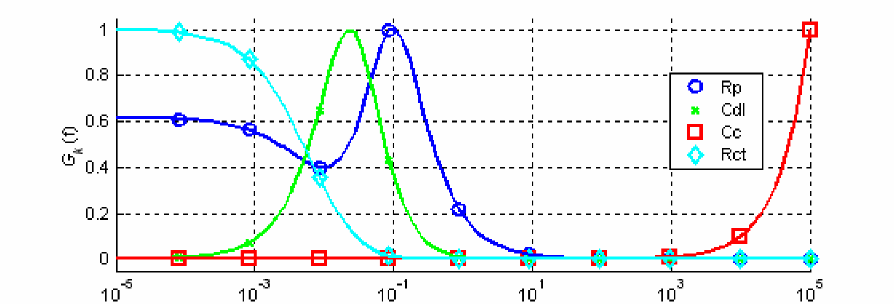

Figure 5. Plot of the objective function for criterion C_3.

By comparing the Fig. 4 and Fig. 5, it can be noticed, that normalization of denominator components has spread the optimal frequencies a bit wider. The R_p optimal frequency is higher (more far from f_{Cdl}), whereas the objective function for parameter R_{ct} descents faster, thus suggesting that a lower frequency should be useful.

Table 2. Vectors of test frequencies for criteria C_1, C_2 and C_3.

Criterion	Frequency optimal for:				Meas. time [s]
	R_{ct}	C_{dl}	R_p	C_c	
C_1	1.85 mHz	21.21 mHz	67.23 mHz	1.22 Hz	601
C_2	124.24 μHz	20.25 mHz	61.31 mHz	98.98 kHz	8114
C_3	66.14 μHz	23.26 mHz	98.72 Hz	98.98 kHz	15171
R_1	100.00 μHz	100.00 mHz	10.00 Hz	100.00 kHz	10010
R_2	*3 points per decade + 1 = 28 test frequencies*				18622

The test frequency vectors for the new criteria has been chosen from a 10 decades wide range of frequencies, in which the impedance spectrum of a nominal coating (model A) changes significantly.

The ε value has been set a 1%.

Additionally, two reference sets of frequencies have been prepared covering the same frequency range. Firstly, the conventional CNLS impedance spectroscopy vector of frequencies (denoted R_2) with 3 points per decade (27 test frequencies) was prepared. Secondly, the vector R_1 covering the frequency range logarithmically with number of test frequencies equal to number of identified parameters has been prepared. Table 2 presents test frequencies generated by criteria C_1, C_2, C_3 and R_1. For the 28-points vector R_2 only the measurement time has been presented for the sake of clarity.

The measurement time for all test vectors were approximated by a sum of periods of all frequencies in a vector, due to fact, that one period is usually sufficient to make the impedance measurement via DFT methods [13].

V. Results and Conclusions

The shapes of the curves being the graphical representation of frequency selection criteria, presented in Fig. 2, 3 and 4 are correlated with “common sense” approach to identification of Beaunier’s model parameters. It can be seen, that the higher the frequency is, the better the capacitor C_c can be identified, as it is shunting rest of the circuit and dominates the impedance. The R_{ct} identification frequency should be as low as possible, in order to minimise current flowing through C_c or C_{dl}, and thus R_p. The C_{dl} capacitor (with higher capacitance then C_c) should be identified at frequency low enough to limit influence of C_c, but higher than the frequency optimal for R_{ct}. The R_p identification frequency is localised at the point where that element influences on both real and imaginary part of impedance, due to C_{dl} and R_{ct} current flowing through it.

Table 3. Standard relative uncertainity of equivalent circuit parameter identification for models A and B with starting values equal to A or B.

Criterion	relative uncertainty [%]			
	R_{ct}	C_{dl}	R_p	C_c
C_1	0.52	1.14	0.35	0.46
C_2	0.61	1.46	0.48	0.83
C_3	0.43	1.03	0.22	0.26
R_1	0.73	1.23	0.35	0.26
R_2	0.23	0.66	0.21	0.15

The wider set of frequencies chosen by criterion C_3 result in prolongation of the measurement time, due to R_{ct} optimal frequency, being the lowest. However, the measurement time is still shorter, as compared to conventional CNLS. If the lowest frequency was limited to 100 μHz, the measurement time reduction would be much more significant.

The relative uncertainties of CNLS parameter identification for designed vectors of test frequencies are presented in Table 3 and 4.

Table 4. Standard relative uncertainity of equivalent circuit D parameter identification with starting values B.

Criterion	relative uncertainty [%]			
	R_{ct}	C_{dl}	R_p	C_c
C_1	2.77	7.10	0.23	0.29
C_2	2.99	8.72	0.27	0.49
C_3	2.43	5.73	0.18	0.20
R_1	4.40	8.83	0.30	0.21
R_2	1.76	4.20	0.16	0.14

The uncertainties of parameter identification for criteria C_1, C_2 and C_3 were very similar for 4 cases: identification of model A and B with CNLS starting values taken from model A and B. All the designed criteria (C_1, C_2, C_3) gave better results (presented in Table 3) than 4 arbitrary chosen frequencies R_1 covering the same frequency range. On the other hand, the accuracy is still worse than with 28-point CNLS.

It seems that, as the real parameters do not differ more than 10 times from starting values, the iterative fitting in LEVM program works well and uncertainty is dependant on a set of frequencies. Between the 3 new proposed criteria, the C_3 gave the best results.

However, if the coating is in stage modelled by circuit C, D or E and we take the nominal (A) starting values the CNLS algorithm does not converge to the proper identification. Very often (up to 92% simulations) the identification with only 4 frequencies was impossible. On the other hand, in these cases even the 28-points CNLS has identified the parameters with high uncertainty.

If the start values were assumed as from model B, the C, D and even E circuits could be identified. Table 4 presents the parameter identification uncertainty for equivalent circuit D with start values B.

To sum up, the results confirm the possibility of accelerating the identification measurement time, although at a cost of accuracy. The criterion C_3 was the most appropriate – it gives better results (lower uncertainty) then proposed C_1, C_2 and 4 equally (in terms of logarithmical scale) distributed frequencies. Moreover, it can be seen, that using the 28-points CNLS test frequencies vector R_2 results in approximately $\sqrt{28/4}$ lower uncertainty then R_1, due to overdetermined set of measurement data.

Another conclusion is that the CNLS method begins to fall when starting values are more then 100 times smaller or greater then actual parameters. In such case, the danger of misconvergence of the CNLS grows when using smaller number of frequencies.

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