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Introducing EIS Circuit Elements in SPICE Simulator Environment

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Abstract: This study introduces hypothetical circuit elements, specifically the Constant Phase Element (CPE) and Zeroth-Order Approximation of a RC Circuit (ZARC), into the SPICE circuit simulation environment to enhance Electrochemical Impedance Spectroscopy (EIS) analysis. EIS, a critical method for understanding electrochemical processes in fields such as fuel cell analysis, corrosion studies, and biomaterials, relies on fitting measured impedance curves to Equivalent Electrical Circuit (EEC) models. However, existing approaches require expert knowledge and significant mathematical effort, limiting automation. By integrating CPE and ZARC into SPICE, this work bridges the gap between EIS analysis and advanced automatic circuit design methodologies, enabling efficient model selection and parameter determination. Experimental results demonstrate the accuracy of the implemented elements through a series of case studies, evaluated using Sheppard's criteria function. This integration marks a significant step toward automated EIS model fitting and optimization, with potential implications for advancing electrochemical and materials research.

Keywords: Electrochemical Impedance Spectroscopy, Circuit Simulators, Hypothetical Circuit Elements, Equivalent Electronic Circuits

Vpeljava Elemementov EIS v SPICE Simlator Vezij

Izvleček:. Ta študija uvaja hipotetične vezne elemente, specifično konstantnofazni element (CPE) in ničelni red približka RC vezja (ZARC), v simulacijsko okolje SPICE za izboljšanje analize elektrokemijske impedančne spektroskopije (EIS). EIS, ključna metoda za razumevanje elektrokemijskih procesov na področjih, kot so analiza gorivnih celic, študije korozije in biomateriali, temelji na ujemanju izmerjenih impedančnih krivulj z modeli ekvivalentnih električnih vezij (EEC). Obstoječi pristopi zahtevajo strokovno znanje in znatno matematično delo, kar omejuje avtomatizacijo. Z integracijo CPE in ZARC v SPICE to delo premošča vrzel med analizo EIS in naprednimi metodologijami samodejne zasnove vezij, kar omogoča učinkovito izbiro modelov in določanje parametrov. Eksperimentalni rezultati potrjujejo natančnost implementiranih elementov skozi serijo študij primerov, ocenjenih s pomočjo Sheppardove funkcije kriterijev. Ta integracija predstavlja pomemben korak proti avtomatiziranemu prilagajanju in optimizaciji modelov EIS, z možnimi vplivi na napredek raziskav na področju elektrokemije in materialov.

Ključne besede: Elektrokemična Impedančna Spektroskopija, Simulatorji Vezij, Hipotetični Elementi Vezja, Ekvivalentna Električna vezja

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1 Introduction

With the rapid growth of electrical storage, electric vehicles, advanced materials and other technological advancements there is also an increasing demand for methods that can offer a quick insight into any ongoing process or material to quickly detect and prevent possible incidents. One of such methods is Electrochemical Impedance Spectroscopy (EIS) which can be used for fuel cell analysis (in order to detect if a fuel cell is starting to deteriorate), corrosion science (analyzing and simulating effects of corrosion on various metals), Bio medics and process control.

The key technique in EIS is measuring the Impedance response and then fitting the resulting curve to a known Equivalent Electrical Circuit (EEC) model. This approach requires some guesswork as the researcher must select one of the known EEC models based on experience and try to fit the model's impedance curve to that obtained during EIS measurement. Each EEC model corresponds to one of the processes encountered in the field (so one model for a perfectly working fuel cell, one for a deteriorating fuel cell, one for a cell with corrosion on connectors etc.). If the model represents a good fit for the measured data, we can assume that we are dealing with the process described with the selected model.

The two major disadvantages of this fitting approach are that it relies on expert knowledge, since only a seasoned observer can reliably select the most appropriate EEC when seeing the measured impedance curve and that the approach also requires a lot of mathematical work to fit the EEC circuit element values to the measured data.

The field of automatic analog circuit design on the other hand offers several methods that could be used to automate this procedure and reduce the required level of expertise. Several advanced techniques could potentially also determine the correct values of EEC elements. All these techniques, however, face a problem – EEC circuits consist of several circuit elements (Constant Phase Element, ZARC) which do not exist in circuit simulators used by these approaches. These elements don't have a direct physical equivalent in a circuit and are mostly represented by their transfer function. For the purposes of this article we will refer to these elements as hypothetical circuit elements to discern from those already implemented in circuit simulators.

The aim of the research presented in this article is to correct this by introducing these hypothetical elements into SPICE circuit simulator and thus enabling future research into combining automatic circuit design techniques with the EEC circuits obtained from EIS.

2 Current state of EIS

Current EIS techniques rely on a combination of Circuit Description Code (CDC) [1], circuit model selection and mathematical modelling techniques such as the Levenberg–Marquardt Algorithm.

2.1 Electrochemical Impedance Spectroscopy

Electrochemical Impedance Spectroscopy (EIS) is a powerful analytical technique used to study the electrical properties of materials and electrochemical systems. By applying a small alternating current (AC) signal over a range of frequencies to an electrochemical system and measuring the resulting voltage response, EIS provides insights into the impedance characteristics of the system. This technique is invaluable for characterizing the complex behavior of electrochemical reactions, interfaces, and materials. EIS is particularly useful in understanding processes in fuel cells, batteries, corrosion studies, and other electrochemical devices.

In the context of fuel cells, EIS is employed to analyze the performance and degradation mechanisms of these energy conversion devices. By applying EIS to fuel cells, researchers can evaluate the impedance at different frequencies to identify various components of the cell's resistance, including ohmic resistance, charge transfer resistance, and mass transport resistance. This allows for a deeper understanding of the fuel cell's efficiency, degradation, and overall performance, facilitating improvements in design and operation.

2.2 Equivalent Electric Circuits and Circuit Description Code

The core of EIS consists of selecting a known circuit and seeing if it is possible to match its impedance curve to measured data.
These circuits are often described using the Circuit Description Code (CDC) which describes circuits using their elements (resistor as R, capacitor as C etc.) and parenthesis to describe their relation [1]. A CDC without parenthesis would describe all elements being in series. Parenthesis denotes elements that are parallel to each other.

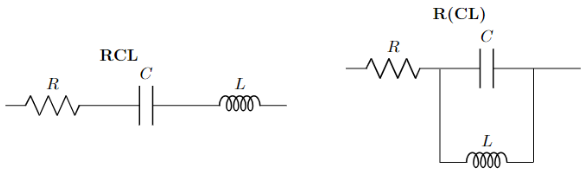

Figure 1: CDC example for a simple serial circuit (left) and a circuit with parallel elements(right)

Each EEC is therefore described with the appropriate CDC code. Once selected it offers a mathematical model to be fitted to the input curve. In the case of a RCL circuit the fitter would have to determine the correct resistance (R), capacitance (C) and inductance (L) of the circuit. One should note that for the purpose of this article, we did not add any additional noise (Gaussian or any other model) since we were aiming to create a circuit model that is a perfect fit to the mathematical model of our hypothetical elements.

2.3 Notable EEC Components and their usage in modelling

While there are many different components used in EEC two of them tend to stand out and are used frequently in fuel cell analysis.

2.3.1 Constant Phase Element (CPE)

The CPE element [2] is used for modelling several simple chemical processes in fuel cells such as porous electrodes, the effect of surface roughness of electrodes, etc. CPE therefore simulates non-ideal capacitive behavior and deviations from standard capacitance. The mathematical model of the element is given by equation 1:

$$Z_{\mathrm{CPE}}(\omega) = \frac{1}{Q(j\omega)^n} \tag{1}$$

where Q is the CPE constant, ω is the angular frequency and n the CPE exponent ($-1 < n \leq 1$), which describes the deviation from ideal capacitive behavior. A CPE element with n=1, would therefore have the same frequency response as an ordinary Capacitor. One should note that with fuel cells the value of n is usually between 0.5 and 1. At 0.5 the CPE becomes the Warburg element that is often used to study diffusion. In CDC the CPE element is denoted with the symbol **Q**.

2.3.2 Zeroth-Order Approximation of a RC Circuit (ZARC)

The ZARC element [3] is used for simulating impedance response with multiple RC-like behaviors and complex impedance spectra. The impedance model of this element is given by equation 2:

$$Z_{\mathrm{ZARC}}(\omega) = \frac{R}{1+RQ(j\omega)^n} \tag{2}$$

where R is the resistance, Q is the CPE constant, ω is the angular frequency and n is the order of approximation, usually between 0.5 and 1, same as with CPE element. In CDC the ZARC element is represented by **(RQ)**. [4]

2.4 Mathematical modelling

Current EIS techniques [5] work by first performing a frequency sweep of the selected process (circuit element, fuel cell or electrode) and measuring its impedance. Once the impedance is known the user must select the EEC model that he/she thinks would fit best.

Once the model is selected, the user must also select the estimated starting values of each element in the EEC model (resistance, CPE constants, orders of approximation…). These are then used with the selected mathematical approach to determine the best fit possible for the selected model and the exact element values at the same time. If the chosen values are not appropriate, the procedure still converges to the correct result but requires more iterations to do so. It can also lead to procedure getting stuck in a local minimum and producing incorrect results. This is a common issue in EIS data fitting, which is why it is desired for the starting values to be close to the optimal values.

Examples of such techniques are reflective Newton type method [6] and Levenberg-Marquardt algorithm [5].

2.5 Sheppard's criteria function

Once the mathematical approximation is complete the results are compared with the original impedance curve using the selected criteria function. Since this is a curve fitting problem there are many possible criteria functions such as RMSE, MSE and R-square. In the field of EIS however it has been estimated that the Sheppard's criteria function [7] [1] performs best for the measured impedance data, especially since it features an additional weight [8] to compensate the difference between imaginary and real data axis. We therefore opted to use this function in our research.
The criteria function is calculated using equations 3 and 4:

$$Shp = \sum\left(w_i\left(Re\left(Z_i^{math}\right) - Re\left(Z_i^{model}\right)\right)^2 + w_i\left(Im\left(Z_i^{math}\right) - Im\left(Z_i^{model}\right)\right)^2\right) \tag{3}$$

$$w_i = \frac{1}{Re\left(Z_i^{math}\right)^2 + Im\left(Z_i^{math}\right)^2} \tag{4}$$

where w_i represents the weight calculated from the input impedance data, Z_i^{math} represents the impedance of the mathematical model (our target) calculated at the i-th frequency point and Z_i^{model} the measured impedance of our SPICE circuit at the same frequency point.

One should note that in cases of real measurements, the Z_i^{math} gets replaced with $Z_i^{measured}$.
The result of the criteria function indicates the goodness of the fit with lower values indicating a better fit. In the event of a perfect fit the result would thus be zero.

2.6 Our alternative approach

The above techniques rely heavily on the user's expertise and experience in addition to requiring a lot of input from him/her (the EEC model and its estimates).

In the field of circuit designs there are a lot of tools that offer a certain level of automatization for circuit design and could potentially be used to help in this example [9] [10] [11] [12]. Such techniques can be based on genetical programming [13], evolutionary computation with a preselected element layout [14] or even grammatical evolution [13] [15] [16] where one can create completely new circuits from the given electrical components.

The only problem is that all such techniques require access to circuit elements which in case of EIS is not possible since CPE and ZARC do not yet exist in most circuit simulators. The only known implementations are approximations using a long chain of RC elements as shown by Lopez [3]. Our aim is to create a single compact element that matches the mathematical model exactly.

We therefore decided to implement and evaluate these elements in the SPICE circuit simulator with the aim of developing an automatic EIS model selection algorithm in the future. This required rewriting some of the SPICE models and exhaustive testing.

2.7 Challenges with implementation of hypothetical elements in circuit simulators

Due to the non-integer exponent of $j\omega$ the transfer functions of CPE and ZARC cannot be modeled with circuit elements like resistors, capacitors, and inductors. The fractional exponent also complicates the simulation of such devices in the time domain where the usual approach is to use convolution with the impulse response of the device. The impulse response can be obtained via inverse Fourier transformation of the device's transfer function. Fortunately, impedance calculations are performed in the frequency domain where the evaluation of CPE and ZARC involves only simple algebraic manipulations of complex numbers.
The internal API of SPICE makes it possible to separately describe the behavior of an element for the time domain and for the frequency domain. The model developer is responsible for making sure these two descriptions are consistent with each other. If one does not intend to simulate a device in time domain, it is sufficient to provide the description of a device for frequency domain only.
Because CPE, ZARC, and all the EIS equivalent circuits are linear the time domain description of CPE and ZARC can be reduced to that of a simple resistor. This description is used only in the computation of the operating point. Because the circuit is linear the linearizations of the elements which are used in the frequency domain

analysis do not depend on the operating point. The operating point computation cannot be skipped thus one has to make sure the simulator computes some operating point that does not even have to reflect any physically meaningful behavior.
Modeling a device in frequency domain involves the computation of its admittance matrix. For CPE and ZARC this matrix reduces to a single element. Its value can be computed by evaluating the inverse of the element's impedance at the frequency provided by the simulator.

3 Adding new elements to SPICE

Our aim was therefore to take the elements required by EIS (CPE and ZARC) and implement them in the circuit simulator of our choosing (SPICE). We also considered alternatives such as Verilog-A [17] but found that we could not use to correctly model the element in the frequency domain.

We have also performed a series of evaluations where we compared the mathematical model (i.e. the ideal) with the results given by the simulator elements. Ideally, they should be as close as possible. In order to remain impartial, we used the same evaluation technique as is used in measuring the fit between the curve and the mathematical model in EIS – the Sheppard's criteria function.

3.1 SPICE

Developed in the late 1970s at the University of California, Berkeley, SPICE (Simulation Program with Integrated Circuit Emphasis) [18] provides a comprehensive environment for modeling and simulation of the behavior of analog and digital circuits.

At its core, SPICE simulates the electrical behavior of circuits by solving the nonlinear differential equations that describe the circuit components and their interactions. It models the various elements of a circuit, such as resistors, capacitors, inductors, diodes, transistors, and operational amplifiers, using mathematical equations that represent their behavior.

One of SPICE's primary functions is to perform a DC operating point analysis, which calculates the steady-state voltages and currents in the circuit. This analysis is essential for understanding the circuit's behavior under constant input conditions. Additionally, SPICE can conduct AC analysis to determine the circuit's response to small-signal variations across a range of frequencies, providing insights into frequency response, gain, and stability.

SPICE has become a cornerstone of electronic design and analysis due to its ability to accurately predict circuit behavior before physical prototypes are built. This capability significantly reduces development time and costs by allowing engineers to test and optimize their designs virtually.

In addition to traditional circuit simulation, SPICE has been integrated into mixed-signal simulation environments that combine analog and digital circuit analysis. This integration allows for comprehensive testing of systems that involve both types of circuits, such as those found in modern microprocessors and digital communication systems.

In research, SPICE facilitates the exploration of new circuit designs, materials, and technologies. Researchers use the simulator to model innovative concepts, test hypotheses, and validate theoretical predictions. This capability accelerates the development of cutting-edge technologies and advances the field of electronics.

3.2 Implementation of CPE and ZARC in SPICE

Adding a new device to the SPICE simulator is a tedious process. Fortunately, XSPICE extensions provide a simple mechanism for adding new elements to SPICE. We used Spice Opus [19] [20] which is based on the original SPICE3 source code with added XSPICE extensions. Spice Opus makes it possible to describe an element by using the XSPICE API. The description is compiled into a dynamic library which is loaded by the simulator at runtime.
In XSPICE one describes the behavior of an element in the frequency domain by specifying the real and the imaginary parts of the transfer functions between element's terminal voltages and branch currents. Because both CPE and ZARC have only two terminals and one branch a single transfer function must be defined. For CPE the transfer function at a given frequency f is

$$H = Q(2\pi f)^n cos(n\pi/2) + iQ(2\pi f)^n sin(n\pi/2) \qquad (5)$$

Similarly, for ZARC we have

$$H = R^{-1}\big(1 + \Omega cos(n\pi/2)\big) + iR^{-1}\Omega sin(n\pi/2) \qquad (6)$$

where $\Omega = (2\pi f\tau)^n$ and i is the imaginary unit.
The element description is provided in two files. One specifies the element's interface (terminals and parameters). The other specifies its inner workings. The latter one is written in an extension of the C programming language. Both files are preprocessed by the XSPICE's model compiler (cmpp) upon which the resulting files are compiled with a C compiler and linked into a dynamic library.
For each candidate circuit PyOpus [21] generates the circuit's netlist comprising its topology and parameter values. The netlist also contains the commands the simulator must execute to produce the circuit's response. After the netlist is created PyOpus invokes the Spice Opus simulator which loads the netlist (Figure 2). The dynamic library with the CPE/ZARC model is loaded into the simulator with the corresponding simulator command (cmload) which is followed by the analysis command. The results are written into a SPICE RAW file from which they are loaded by PyOpus for impedance extraction and cost function computation.

3.3 Simulation and Evaluation

The aim of our experiments was to match the theoretical (i.e. mathematical) circuit elements as closely as possible with simulated circuit elements in SPICE. We first used the mathematical impedance model to create the baseline impedance curve and compared it with the SPICE simulation results.

3.3.1 Input Data – Mathematical Model

For all our experiments we used the mathematical impedance model of the selected circuit. We used the models described by equations 1 and 2 computed at given points in the selected frequency range.
For some of the case studies we also added some Gaussian noise to the calculated impedance curve to see if it would have any effect on the final evaluation.

3.3.2 Output Data – SPICE simulation

Our SPICE simulations used a simple circuit for measuring the desired impedance curve.
The system measured the voltage values at both ends of the subcircuit and then calculated the resulting impedance. We used the AC analysis with a frequency sweep from 10^{-2}Hz to 10^{5}Hz which is the usual range used in EIS measurements.
Our experiment consisted of two circuit files – one for the main circuit (as shown on figure 1) and the other for the element setup we wanted to measure. This enabled us to perform a whole series of experiments by simply swapping out the subcircuit files.

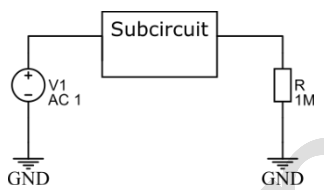

Figure 1: Measuring circuit setup

3.3.3 Evaluation

Since we wanted to be able to compare our results to those presented in other EIS articles, we used the same metric that is used by them – Sheppard's criteria function (see 2.5). The only difference is that we compared the impedance curve gained from the mathematical model (using equations (1) and (2)) with the impedance values obtained from the SPICE simulator. Everything else remained the same – the lower the number the better our SPICE model matched the theoretical mathematical model of the EEC element.

3.3.4 Experiment flow

The main aim of our experiments was to prove that our circuit model matches the theoretical model of each element. Instead of first calculating the model impedance values for each frequency of interest and then comparing the result with the curve obtained from the SPICE element we made a slight modification.
We first ran the SPICE simulation and collected both the impedance values, and the exact frequencies used by the simulator. We then used these points in the mathematical model to ensure a fair comparison.
Lastly, we used the Sheppard's criteria function to get the result.

4 Case studies

We tested three circuits that commonly occur in EIS articles – a single CPE element, a single ZARC element and two serial ZARC elements, combined with a resistor. The last circuit is also often used in Fuel cell EEC. For each circuit, we first calculated the values of the mathematical models using equations 1 and 2. We then created an equivalent subcircuit netlist, ran the SPICE simulation and extracted the impedance measurements. Both the model and the measurements were then used in our final evaluation using the Sheppard's criteria function. One should note that at this stage we used only mathematical models as input since we had to verify the fit of new circuit elements.

4.1 Single CPE element (Q)

The single CPE element had the CPE constant set to 0.01 and the n value to 0.8 with no additional elements in the circuit. As can be seen in figure 2 the model and simulation were a perfect match, with a Sheppard value of $7.1 * 10^{-13}$.

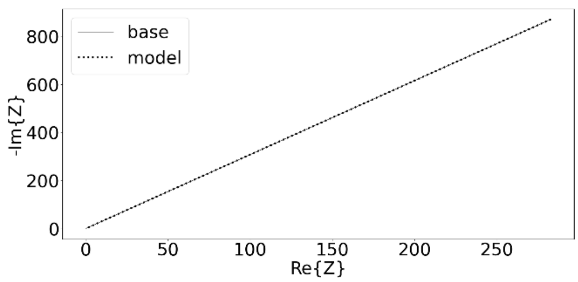

Figure 2: Single CPE

4.2 Sigle ZARC element (RQ)

For the single ZARC element we used resistance value of 50Ω, CPE constant 0.01 and n value of 0.7 and again added no additional elements to the subcircuit. The resulting Sheppard value was $7.09 * 10^{-13}$ with the model and simulation data shown in figure 3.

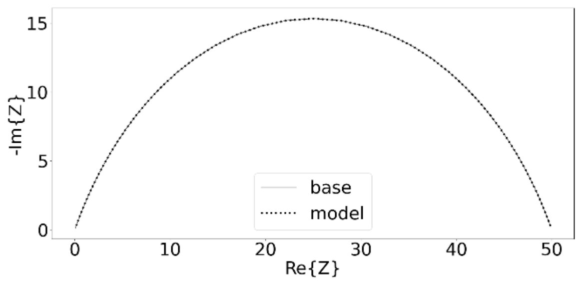

Figure 3: Single Zarc

4.3 Two parallel ZARC elements (R(QR)(QR))

This was by far the most important experiment since there are a lot of cases where fuel cell measurements result in the “double hill” curve which indicates that there should be two ZARC elements involved. The EIS model adds an additional serial resistor, so we adjusted our subcircuit accordingly.
The first evaluation featured ZARC elements with similar characteristics and is shown in figure 4. The circuit featured a serial resistor of 10Ω, first ZARC with (R=50 Ω, CPE constant = 0.01 and n = 0.7) and the second with (R=50 Ω, CPE constant = 0.0001 and n = 0.7). The resulting criteria function value was $2.17 * 10^{-12}$ which again show a good fit.

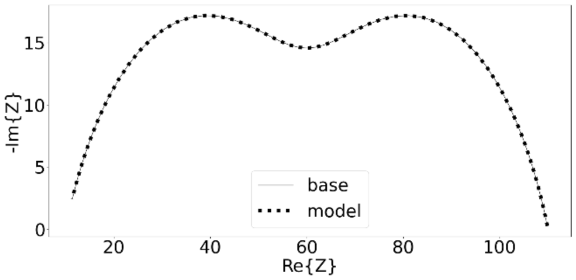

Figure 4: Double Zarc Model

It is however rare to have ZARC elements that are so evenly matched, which is why we performed another evaluation and adjusted the second ZARC element to (R=50 Ω, CPE constant = 0.01 and n = 0.45). The resulting fit is shown in figure 5. The value of the criteria function was $2.54 * 10^{-12}$ which means that the fit remains good despite a noticeable difference from the original curve shown in figure 4.

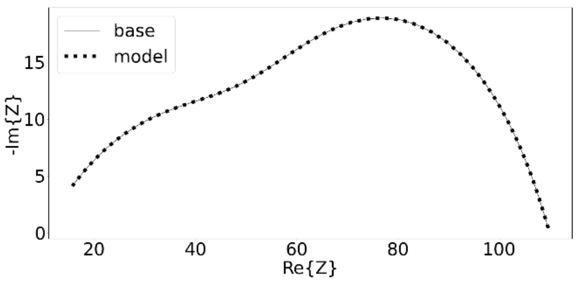

Figure 5: Two ZARC with dissimilar characteristics

5 Conclusions and future work

The results presented in this work show that we have successfully implemented two theoretical circuit elements (CPE and ZARC) in the SPICE circuit simulator. This is one of the first known implementations of such elements in SPICE.

This implementation allows a completely new approach to EIS impedance fitting since we can now experiment with automatic circuit design techniques which were previously inaccessible in the field of EIS. It also does not impact the performance of SPICE since we only added additional models to its circuit elements library.

For our future work we plan on trying to modify our existing evolutionary methods [16] [22] to include the new elements and try to select the correct EEC model based on the input impedance curve. Should this prove successful we can also attempt to correctly determine the circuit element values using circuit optimization techniques [23].

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7 Conflicts of interest

The authors declare no conflict of interest.
The founding sponsors had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript, and in the decision to publish the results.

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