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Conference Paper *in* Journal of Computational and Nonlinear Dynamics · June 2008

DOI: 10.1109/MELCON.2008.4618411 · Source: IEEE Xplore

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Modeling Electrochemical Double Layer Capacitor, from Classical to Fractional Impedance

Rodolfo Martin, Jose J. Quintana, Alejandro Ramos, and Ignacio de la Nuez

***Abstract*—The application of the fractional calculus for modeling electrochemical double layer capacitors is a novel way to get simpler and precise models. This paper provides a summarized report of several models of Electrochemical Double Layer Capacitors (EDLC). From classical models based on simple structures composed by networks of passive elements (RLC), to models which involve the fractional behavior of EDLC. The fractional models are based on one hand by a function with fractional poles and zeros and for other one by electrochemical elements (Warburg, Bounded Warburg, Havriliak-Negami). On using the impedance spectroscopy method, experimental results for different EDLCs have been obtained. Coming to the conclusion that the fractional models are fitted better than the classical models.**

***Index Terms*—Fractional calculus, electrochemical double layer capacitors, Impedance spectroscopy, Modeling.**

I. Introduction

This paper provides a summarized report of several models of Electrochemical Double Layer Capacitor (EDLC). From classical models based on simple structures composed by networks of passive elements (RLC), to models which involve the fractional behavior of EDLC.

EDLC is a technology, which has emerged with the potential to enable improvement in energy storage [1–3]. EDLCs behavior is like conventional capacitors, but they utilize higher surface area electrodes and thinner dielectrics to achieve greater capacitances. This imply energy densities greater than those of conventional capacitors and power densities greater than those of batteries.

Electrochemical double layer capacitors typically have energy densities that range from 300 times that of the largest conventional capacitors, to approximately two tenths of that of the lowest density batteries. However, their power densities are typically 10 times that of most batteries. By offering high power and energy densities coupled with low equivalent series resistance (ESR), EDLC bridges the gap between batteries and conventional capacitors [1].

EDLCs are similarly known as supercapacitors or ultracapacitors stores energy electrostatically by polarizing an electrolytic solution. Though it is an electrochemical device there are no chemical reactions involved on its energy storage mechanism. This mechanism is highly reversible, allowing the ultracapacitor to be charged and discharged hundreds of thousands to even millions of times.

Electrochemical double layer capacitors are used in automotive applications for electric (including hybrid electric) vehicles and as supplementary storage for battery electric vehicles, in electronic devices like VCR circuits, CD players, computers and other electronic equipments.

It is quite important to obtain an accurate model of EDLCs, because is necessary in order to get high performances in different applications, in which EDLCs are involved. Impedance spectroscopy analysis is the typical way in order to carry out the identification of several electrochemical systems, like EDLCs. It is an analysis in frequency domain. From this analysis it has been observed that classical models, based on RLC networks, need to be very complex in order to achieve an accurate identification. Therefore in the electrochemical field is used models with fractional elements, to obtain better results in the identification process.

The aim of this paper is to show that is able to obtain better accuracy with less parameters using fractional models than using traditional models.

This paper is organized as follows: section II describes a survey of fractional calculus; section III shows a brief description of EDLCs; section IV expounds the diferents EDLCs mathematical models; section V displayes the results; and finally in sections VI the conclusions are presented.

II. A survey of fractional calculus

The idea of non-integer derivates is as old as regular calculus. Fractional calculus has been used for modeling different physical phenomena [4] and in control theory [5–8].

We can notice systems in nature with fractional behaviour, but many of them with a very low fractionality. This way, a technique for partially solving a family of diffusion problems is proposed [9]. A generalized diffusion equation fractional Fokker-Planck equation has been exposed in order to model different anomalous diffusion phenomena [10–12].

There are many examples of real applications of fractional calculus: for the control of electrical machines [13–15], control of power converters [16], in mechanical systems (dynamic models that governs the relaxation of water on a porous dyke) [17], fractional models to describe viscoelastic materials that are used in shock absorbers and flexible arms [18–20], electromagnetics field fractional models [21], several analyses of different fractional dynamics of systems with long-range interaction [22, 23], in the theory of dielectric relaxation [24–27], identification techniques based on fractional electrochemi-

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cal dynamics in oscillographic polarograms, fruits, vegetables, and fuel cell [28–30], and battery state of charge estimation [31].

A typical example of a fractional order system is the voltage-current relation of a semi-infinite transmission line, this being a diffusion phenomenon, which is defined by fractional calculus by different authors, like Olivier Heaveside in 1893 and others [32–34]. Order derivative of the conventional model of RC transmission line is high and involves many parameters. The fractional derivative approach is solely defined in terms of a unique parameter, the fractional order of the derivation. It is able to conclude that fractional calculus is appropriate for systems, which usually are expressed by means of distributed parameters.

It seems important to talk about fractal interpretation of several systems, which are modeled by means of fractional calculus [35, 36]. Especially the characterization of the rough surfaces and interfaces, which has played an important role in understanding the anomalous behavior of these systems. For example a pore fractal, a dense object in which there exist a distribution of holes or pores with a fractal structure. Since it is one of the most interesting issues to estimate accurately real active area in an electrochemical system.

III. DIFFERENCES BETWEEN CAPACITORS AND EDLC

In this section a brief description of the EDLC structure and a comparison with conventional capacitors is presented.

A. Internal construction

An EDLC has two non reactive porous electrodes immersed in a electrolyte, with a separator between the electrodes that permits the movement of the ions through it.

The energy is stored by charge separation in an electrochemical double layer, formed at the electrode/electrolyte interface, fig. 1. The thickness of the double layer depends on the concentration of the electrolyte and on the size of the ions and is in the order of 0.5 to 1 nm for concentrated electrolytes. On the other hand, electrodes are fabricated from high surface area porous material, having pores of diameter of the nanometer range obtaining a specific surface area of about 500-2000 m^2/g and specific capacitances using carbon electrodes of 75-175 F/g for aqueous electrolytes and 40-100 F/g using organic electrolytes [1, 2].

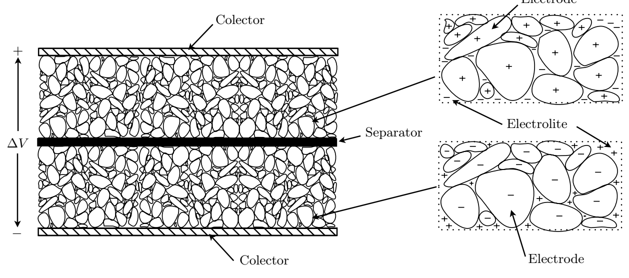

Fig. 1. Internal structure of EDLC

The ions displaced for charging the EDLC are transferred between the porous electrodes by diffusion through the electrolyte.

B. Electrical characteristics

Differences between conventional capacitors and EDLC are not only in their internal structure but also in their electrical characteristics. Figure 2 shows the conventional Nyquist plot of the capacitor impedance like a resistor in series with an ideal capacitor.

$$Z(jw) = R + \frac{1}{j\omega C} \tag{1}$$

Westerlund and Ekstam [37] proposed that the real capacitor has a fractional behavior given by,

$$Z(jw) = R + \frac{1}{(j\omega)^{\alpha} C} \quad 0 < \alpha < 1 \tag{2}$$

where α is close to 1.

In EDLC the diffusion phenomenon in the electrolyte and the size of the electrodes pores are very important. So, when the frequency is risen, the number of active porous layer accessible are reduced, diminishing therefore the resistance and the capacitance [3]. This phenomenon give a fractional behavior to EDLC in a frequency band called Warburg region.

Figure 2 shows that the behavior of EDLC at low frequencies is similar to the capacitors. But in the Warburg region, the equation that relates impedance and frequency is better using the operator $\sqrt{jw}$ than using the traditional operator jw [38].

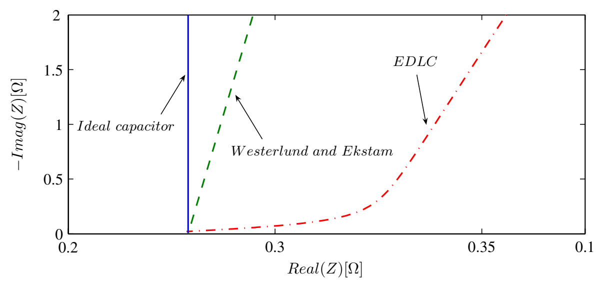

Fig. 2. Nyquist diagram of a capacitor (ideal and Westerlund-Ekstam) and EDLC.

IV. MODELING OF EDLC

The experimental data obtained to measuring the EDLC's must be identified with a model. Then, the mathematical model will be compared with the experimental data to obtain a minimum error and with the minimum possible number of parameters. In this section are presented three different ways of modeling EDLC's in frequency domain.

A. Classical structural modeling

A structural model assumes that the mathematical impedance model can be represented directly in the frequency domain as a construction, consisting of elements. They are connected under different laws in accordance with the real behavior of the system under investigation. Every element describes a single physical process, taking place in the impedance object.

The EDLC's can be modeled as electric networks based in resistors and capacitors [39, 40], fig. 3. It is obtained excellent results at low frequency by means of Maxwell, Volgt or Ladder topologies [41], fig 4. Nevertheless, at medium frequencies, over $400mHz$, the fractional models achieves, with less parameters, an much better fitted than classical model.

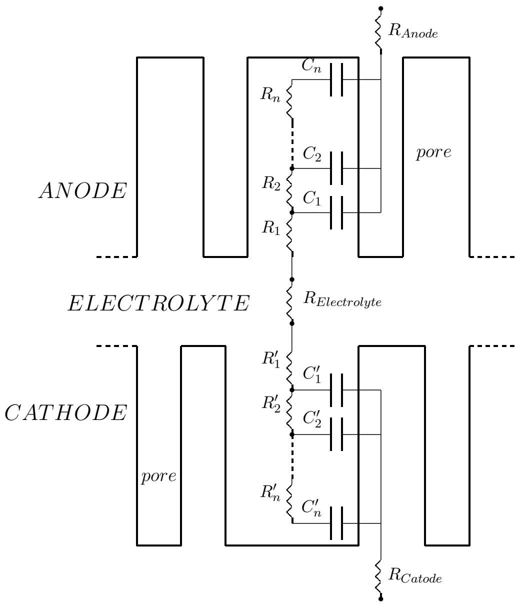

Fig. 3. EDLC model based on resistors and capacitors.

B. Model based on fractional poles and zeros

The equation proposed to model the EDLC by means of fractional poles and zeros [42] is

$$Z(j\omega) = R_s + k\frac{\left(1 + j\frac{\omega}{\omega_0}\right)^{\alpha}}{(j\omega)^{\beta}} \tag{3}$$

This equation is deduced from figures 5 and 6. Where R_s is related to the resistance at 100 Hz in the Nyquist diagram, k is a parameter inversely proportional to the capacitance. The other parameters are deduced from the bode diagram of $Z - Rs$, where β and $\beta - \alpha$ are related to the impedance phase at $50mHz$ and $100Hz$ respectively, and ω_0 is the frequency in which the phase change.

C. Fractional structural modeling

It is difficult to represent electrochemical processes that take place in EDLC. To describe them better it is necessary to define the frequency dependent element - called electrochemical elements - that are developed especially for describing some of these processes and corresponds to an assembly of resistor and capacitors arranged as a fractal array [43]. In this paper

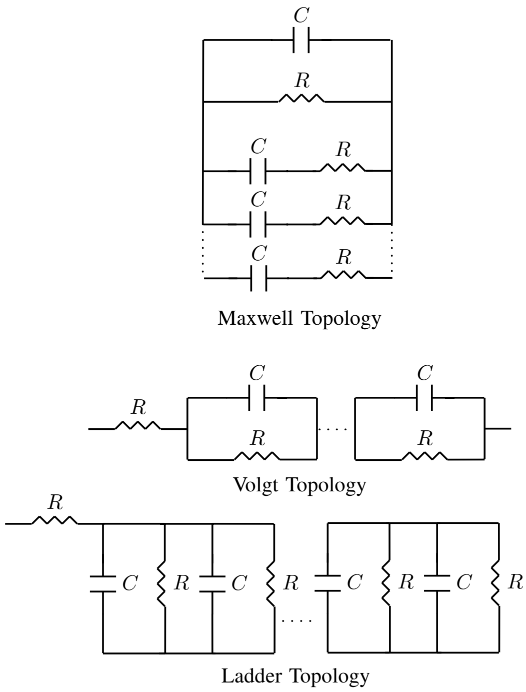

Fig. 4. Maxwell, Volgt and Ladder topologies.

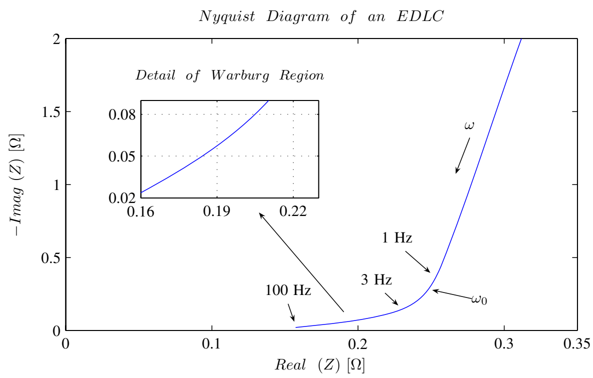

Fig. 5. Nyquist diagram of an EDLC

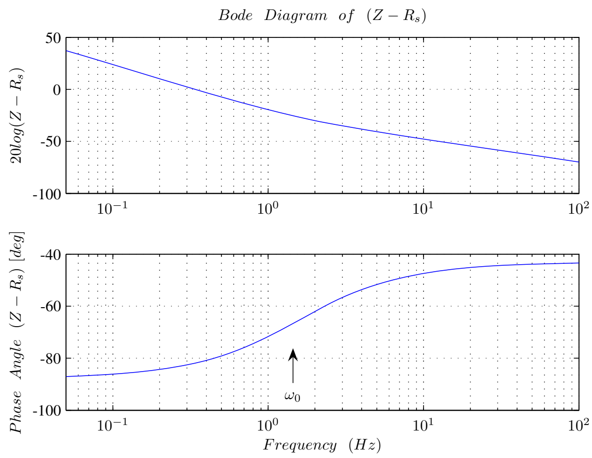

Fig. 6. Bode diagram of $(Z - R_s)$

some electrochemical elements are shown, and whose transfer functions are represented by the following equations:

$$W(j\omega) = \frac{Z_0}{\sqrt{j\omega}} \quad (4)$$

$$O(j\omega) = \frac{Z_0 \coth(B\sqrt{j\omega})}{\sqrt{j\omega}} \quad (5)$$

$$H(j\omega) = \frac{1}{j\omega(C_0 - C_\infty)\left[1 + (j\omega\tau_0)^\mu\right]^\Phi} \quad (6)$$

Warburg impedance, $W(j\omega)$ [44], has been introduced in the impedance description of a linear semi-infinite diffusion, which depends on frequency, and of the potential of disturbance. The Warburg element is a function of only one parameter, Z_0.

Bounded Warburg element $O(j\omega)$ has been introduced in impedance description of linear diffusion in a homogeneous layer with finite thickness [45]. This function characterizes through two parameters, Z_0 and B.

$H(j\omega)$ is the most versatile function in the frequency domain and is due to Havriliak and Negami [46]. This function generates broad, asymmetric curves for impedance-vs-*log w* that are skewed to high frequencies. It is characterized with 4 parameters. The parameter μ $(0 < \mu \leq 1)$ is a measure of the broadness of symmetric impedance curve and Φ $(0 < \Phi \leq 1)$ is the shape parameters of the asymmetric impedance curves.

Figure 7 shows two topologies of four and five parameters, which have been used in order to minimize the number of parameters.

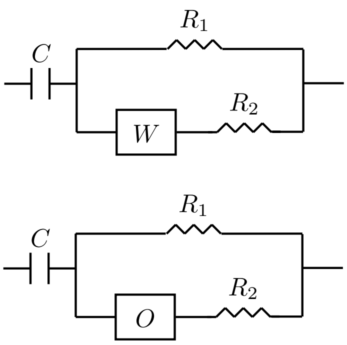

Fig. 7. Fractional models, based on Warburg and Bounded Warburg functions.

Finally, a model of 9 parameters [47] is presented in the figure 8 which achieves an excellent fitting on the over-all frequencies.

V. Results

In this section is shown a briefly compilation of results of the different models, exposed in this paper.

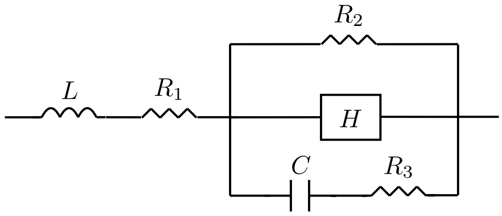

Fig. 8. Fractional models, based on Havriliak-Negami function.

A. Model based on fractional poles and zeros

The results shown are for an EDLC manufactured by EPCOS, with a capacitive value of $5F$ and a voltage of $2.3V$. Figure 9 displays the Nyquist diagram of the measured data in solid line, and the model proposed using the equation 3 in dashed lines, in frequency band from $50mHz$ to $100Hz$. The equation parameters obtained for the EDLC are shown in table I.

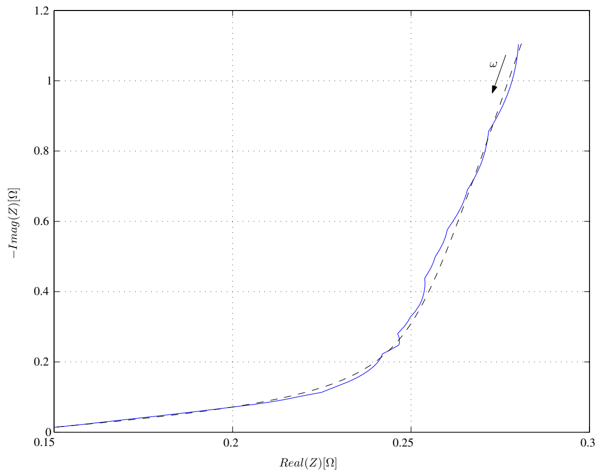

Fig. 9. Experimental data in solid line and model proposed.

TABLE I
VALUES FOR EQUATION 3.

R_s	k	ω_0	α	β
0.1351	0.3435	1.5679	0.5	0.9772

The results obtained with this model are good, and although this model needs for its characterization of 5 parameters shown in Table I. It is demonstrated for a family of EDLCs that the parameters α, β and ω_0 are constant and R_s and k are only function of the capacity value [48].

B. Fractional structural modeling

The results shown are for an EDLC manufactured by PANASONIC, with a capacitive value of $50F$ and a voltage of $2.3V$.

In order to obtain the optimal parameters the mathematical model was compared with the experimental data, in frequency domain, and it has been used a weight function given by (7).

$$\sigma = \sqrt{\frac{\sum_{i=1}^{N}\left(Z^R_{exp_i} - Z^R_{calci}\right)^2 + \left(Z^I_{exp_i} - Z^I_{calci}\right)^2}{N-1}} \quad (7)$$

Where N is the number of samples, Z^R the real part of impedance Z, Z^I the imaginary part of impedance Z, Z_{exp} experimental data, and Z_{calc} calculated impedance.

In the table II can be noticed the results of the weight function for the structural models. The classical models need 7 parameters in order to obtain the results showed. A greater number of parameters do not get better the fitting.

It is observed that the fractional order models of fig. 7 have, in general, fewer parameters and low values of σ are obtained.

Finally a model of 9 parameters displayed in fig. 8 achieves an excellent fitted on the overall frequencies.

TABLE II
VALUES FOR FRACTIONAL STRUCTURAL MODELING II.

	$Classical$	W	O	H
$\sigma \times 10^6$	860	302	73.8	7.36

VI. CONCLUSIONS

In this paper has exposed a summarized report of several models of Electrochemical Double Layer Capacitors (EDLC). From classical models based on simple structures composed by networks of passive elements (Maxwell, Volgt, Ladder), to models which fractional behavior. The fractional models are based on one hand by a function with fractional poles and zeros and for other one by electrochemical elements (Warburg, Bounded Warburg, Havriliak-Negami). On using the impedance spectroscopy method, experimental results for different EDLCs have been obtained, for $5F$ EDLC manufactured by EPCOS and for $50F$ manufactured by PANASONIC. Coming to the conclusion that the fractional models are fitted better than the classical models.

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