



Ragone Plots for Electrochemical Double-Layer Capacitors

Rafael Vicentini,^[a] João Pedro Aguiar,^[a] Renato Beraldo,^[a] Raissa Venâncio,^[a] Fernando Rufino,^[a] Leonardo M. Da Silva,^{*[b]} and Hudson Zanin^{*[a]}

The Ragone plot (RP) compares devices in energy and power density characteristics, helping engineers decide on a specific energy storage system for technological applications. RP would contrast the performance of several technologies together under similar boundary conditions. Unfortunately, that is not what is happening for *electrochemical double-layer capacitors* (EDLCs) because conditions for calculating energy and power are not standardized in the literature, resulting in severe discrepancies in the obtained values. With this widespread

misperception, RP may become an unreliable tool for characterizing EDLCs. An intervention in this vital issue now becomes necessary. To put action on that, a rational strategy for dealing with measurements needed for EDLCs and calculations to prepare a reliable RP is reported here. This manuscript presents a step-by-step procedure to provide a correct RP. Also, simple theoretical demonstrations of how to get each fundamental equation are presented. In general, this work contributes as a guide to obtain reliable RPs for EDLCs.

1. Introduction

1.1. The Ragone plot and its some issues in the study of small-scale laboratory prototypes

Capacitance,^[1] *working voltage window* (WVV),^[2] and *equivalent series resistance* (ESR or R_s)^[3] are the most important fundamental features of *electrochemical double-layer capacitors* (EDLCs). Energy and power densities can be summarized as Ragone plot (RP), which is nothing but a chart in cartesian coordinates with energy and power values normalized by the pertinent, but arbitrary, masses of the entire device or its major components.

The RP was presented for the first time at an automotive engineers conference in 1968.^[4] Since that time, RP has been evolving over the years, presenting energy and power densities on a logarithmic scale to compares the overall behavior exhibited by different energy storage devices such as EDLCs, batteries, fuel cells, etc. Thenceforth, researchers worldwide have been using the RP to present their devices' importance and relevance to the scientific community and industrial sector. Although it seems a well-established topic, for the last ten years, the literature regarding EDLCs has been presenting

different methods to measure and plot the energy and power data in RPs.^[5–258] Figure 1a, b depicts the equations employed for energy and power calculations found in a literature survey using the terms EDLC, supercapacitor, energy, and power as the keywords. The most 25 cited articles were considered to construct Figure 1. Although excellent books and reviews dealing with Ragone's theory are available,^[259,260] only a few

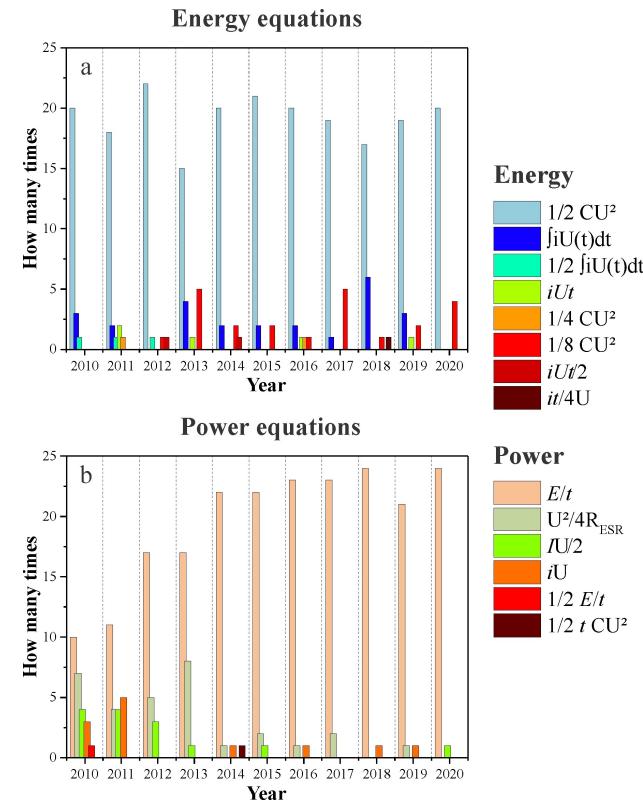


Figure 1. a, b) The bar charts showing a review of the methods used to obtain RPs in the last ten years. The most cited 25 articles for each year were chosen.

[a] R. Vicentini, J. P. Aguiar, R. Beraldo, R. Venâncio, F. Rufino, Prof. Dr. H. Zanin
Advanced Energy Storage Division,
Center for Innovation on New Energies, Manufacturing Group,
University of Campinas,
School of Electrical and Computer Engineering
Av Albert Einstein 400
13083-852 Campinas, SP, Brazil
E-mail: hzanin@unicamp.br
[b] Prof. Dr. L. M. Da Silva
Department of Chemistry, Laboratory of Fundamental and Applied Electrochemistry,
Federal University of Jequitinhonha e Mucuri's Valley
Rodovia MGT 367, km 583, 5000 Alto da Jacuba
39100-000 Diamantina, MG, Brazil
E-mail: lsilvamorais@hotmail.com

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authors have been used the appropriate methods in the case of EDLCs. This remarkable discrepancy about a correct understanding involving RPs has caused severe inconsistencies in the scientific literature. In light of these considerations, some actions are necessary to establish a sensible guide for constructing RPs for EDLC systems.

The literature's inconsistencies arise from the equations employed to calculate the intensive parameters (e.g., areal, volumetric, and gravimetric quantities) used as descriptors of the overall electrochemical performance of EDLCs. For proper normalization, the use of small-scaled electrodes causes considerable deviations in the calculated electrochemical parameters due to the uncertainties in the masses less than *ca.* 1 mg, for example. The experimental scenario is further complicated when a small mass loading characterizes small-scaled electrodes (e.g., less than 1 mg cm^{-2}) allied to a reduced average thickness (e.g., less than 100 μm). Another problem is due to the essential differences between the primary, secondary, and tertiary current distributions on small and large

electrodes.^[261,262] This problem is even more complicated in cells composed of highly porous electrode materials, as is the case of those used in different EDLCs. Therefore, the use of tiny devices (e.g., coin cells) in bench-scale experiments should be avoided since the obtained electrochemical findings do not permit extrapolations for real-scale devices of technological interest.

A wrong evaluation of the energy storage capabilities is also obtained when the normalization procedure does not include the cell encapsulation mass (e.g., cell-housing structure).^[263] In this sense, from a practical (technological) perspective, the RP-based analysis should only be considered using the overall's mass device. However, this assertion touches a delicate topic needing further discussion by the scientific community since most of the literature reports are based on small (bench-scale) electrochemical devices. The goal here is to propose some basic guidelines for authors to obtain meaningful findings during the study of EDLCs using more appropriate sizes for the bench-scale systems. A literature



Rafael Vicentini is a Ph.D. student and Lab Technician at the University of Campinas on the development of energy Storage devices (supercapacitors) at Advanced Energy Storage Division. In 2019, he was awarded the "Development and Innovation Award" in the master's category by the Center for Petroleum Studies (CEPETRO). His research has emphasis on carbon-based materials, electrochemical analysis and Raman characterizations under dynamic conditions.



João Pedro Aguiar is currently an M.Sc. student on the development of energy storage devices in the Advanced Energy Storage Division at the University of Campinas. His research is focused on investigating the effects of ion dynamics in supercapacitor electrodes under compression and low-temperature conditions.



Renato Beraldo is Ph.D. student in electrical engineering at State University of Campinas at GFET based biosensor. His research intents to the development of biosensors with graphene and nanoparticles.



Raissa Venâncio is a M.Sc. student on the development of supercapacitors in the Advanced Energy Storage division at University of Campinas. Her research focuses on electrolytes characterization and analysis of evolved gases in different supercapacitors assemblies under dynamic conditions.



Fernando Rufino is a Ph.D. Student in Electrical Engineering at University of Campinas on the development of Energy Storage devices at Advanced Energy Storage Division. His research is focused on characterization of Graphene Electrodes by AFM analyses.



Leonardo M. Da Silva, Ph.D., is a Professor of Physical Chemistry and Electrochemistry at the UFVJM/Brazil. He is a former director of the Faculty of Exact Sciences (UFVJM). He is the head of the Laboratory of Fundamental and Applied Electrochemistry (LEFA/UFVJM) founded in 2009. He was awarded the "Oronzo and Niccolò De Nora Foundation Young Author Prize (2004)" by the International Society of Electrochemistry (ISE/Lausanne). His current research interests are in the field of Energy Storage Systems.



Prof. Hudson Zanin works with the University of Campinas on the development of Energy Storage devices at the Advanced Energy Storage Division. He has been working on the synthesis and characterization of advanced materials as well as the characterization of electrodes under dynamic polarization conditions.

survey reveals a remarkable lack of discussion about this critical topic.

1.2. General scheme to correctly obtain the Ragone plot for EDLCs

After the controversial topic related to normalization to obtain gravimetric quantities mentioned before, it is now discussed how to prepare RP for EDLCs. Figure 2 shows a flow chart to guide the basic experiments and the correct interpretation of the electrochemical findings.

Firstly, to construct an RP, it is necessary to obtain the following equation:

$$E_L = \frac{E_{\max}}{2} \left(1 + \sqrt{1 - \frac{P_L}{P_{\max}}} \right) \quad (1)$$

where E_L and E_{\max} stand for load energy, while P_L and P_{\max} are the load and maximum power, respectively. Equation (1) will be demonstrated later on in this work.

Energy and power are defined as follows [Eqs. (2), (3)].^[264]

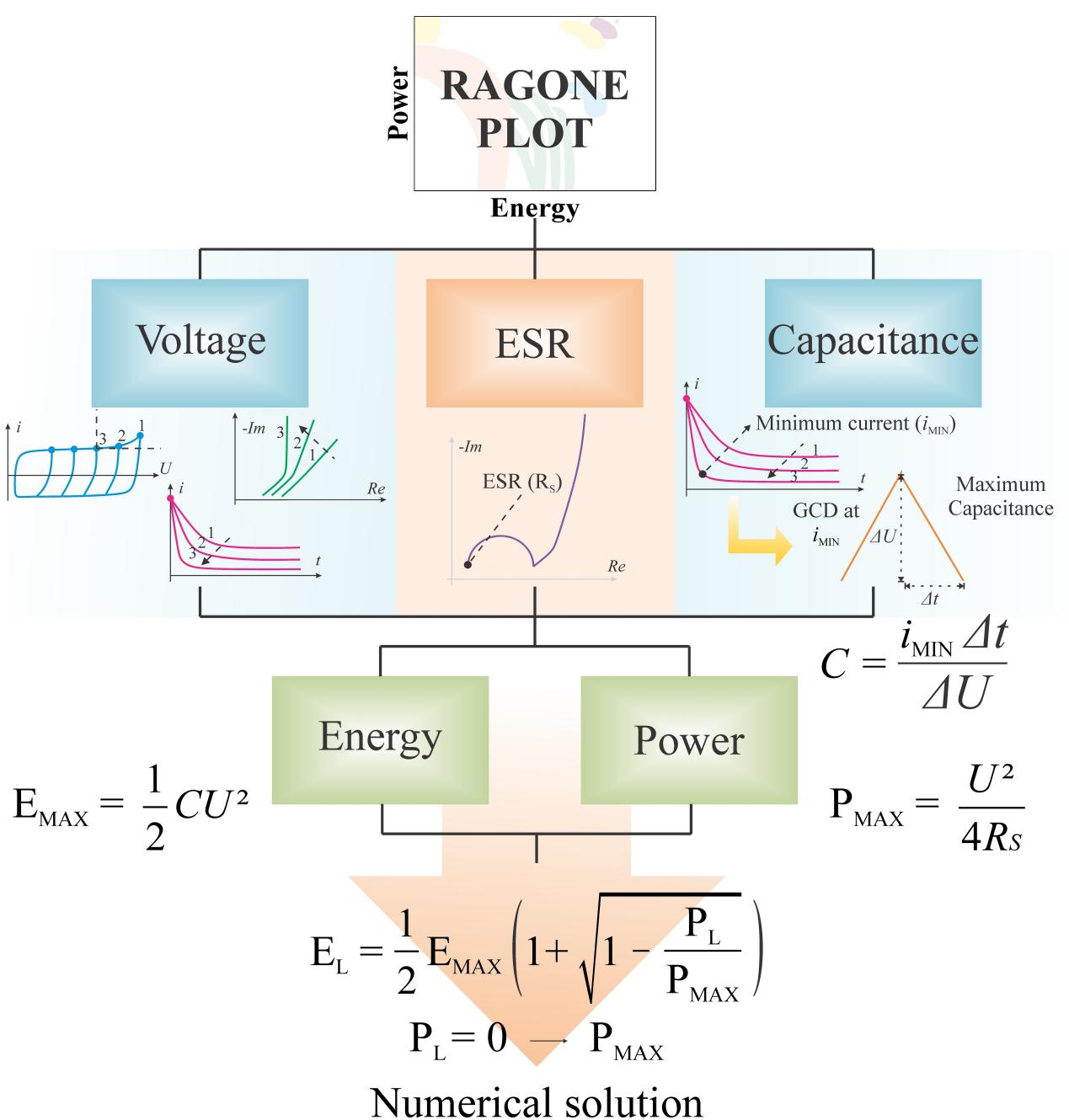


Figure 2. Flow chart with the schematic step-by-step methodology used to construct the RP for EDLCs.

$$E_L = \frac{C_{EDL} U_c^2}{2} \quad (2)$$

$$P_L = \frac{U_c^2}{4R_s} \quad (3)$$

It can be advanced that to obtain the RP one first needs to identify the maximum cell voltage (U_c) where the electrolyte is stable (e.g., WVV), the R_s including all ohmic contributions, and the *maximum capacitance* (C_{EDL}) for the corresponding WVV. With both U_c and C_{EDL} parameters available, the *maximum stored energy* (E_{max}) can be determined. For the sake of identifying U_c , one needs to combine a low scan rate (e.g., $\nu \leq 10 \text{ mVs}^{-1}$) experiment using *cyclic voltammetry* (CV) allied to the use of the *electrochemical impedance spectroscopy* (EIS) and *single-step chronoamperometry* (SSC) techniques as recently reported.^[2] It is worth noting that for highly porous electrodes with great accessibility of hierarchically interconnected micro-pores ($d \leq 2 \text{ nm}$) the scan rate can be even lower to attain the appropriate electrochemical response (e.g., $\nu \leq 1 \text{ mVs}^{-1}$). The same rule applies to host porous material characterized by a solid-state diffusion of intercalating ionic species.

1.3. Cyclic voltammetry and working voltage windows: thermodynamics and kinetics aspects

Briefly, for unknown (as-assembled) electrochemical cells, the CV experiments must be accomplished from a relatively low voltage (U_c) interval of *ca.* 0.2 to 0.4 to ensure initial conditions where the electrolyte is stable. To be practical, the voltage increase can be made in small increments of *ca.* 0.1 V with a low scan rate (e.g., $\nu \leq 10 \text{ mVs}^{-1}$). Even so, the voltammetric profile can hide the presence of electrolyte decomposition. In this sense, it is highly recommended to double-check the data using a very slow scan rate of *ca.* 1 mVs^{-1} to ensure the absence of anodic and/cathodic current tails. The well-known *RC*-model for ideal EDLCs predicts a *transient capacitive voltammetric current* (I_c) at low voltages followed by a *current plateau* at higher voltages according to the following equation [Eq. (4)]:

$$I_c = Cv \left[1 - \exp\left(\frac{-\Delta U}{R_s Cv}\right) \right] \quad (4)$$

However, in the presence of electrolyte decomposition (e.g., water-splitting), the voltammetric profile might reveal upward/downward curved borders (e.g., current tails) for the anodic/cathodic scans, respectively. For the sake of simplicity, in the case of the *anodic* (positive) voltage scan, the overall current, including capacitive and resistive contributions, under high-field approximation conditions (e.g., $U - U_{0(OER)} \geq 70 \text{ mV}$), is given as follows [Eq. (5)]:

$$I = I_c + I_f = Cv \left[1 - \exp\left(\frac{-\Delta U}{R_s Cv}\right) \right] + I_{0(a)} \exp\left[\frac{\alpha_a F(U - U_{0(OER)})}{RT}\right] \quad (5)$$

where $I_{0(a)}$ is the *anodic exchange current density* measured at $U = U_{0(OER)}$, α_a is the anodic charge-transfer coefficient, R is the universal gas constant, and T is the absolute temperature.

Accordingly, the *cathodic* (negative) voltage scan counterpart of the voltammetric curve (e.g., $|U - U_{0(HER)}| < 70 \text{ mV}$) for the reversal scan ($\nu < 0$) is given by Eq. (6):

$$-I = I_c + I_f = -Cv \left[1 - \exp\left(\frac{\Delta U}{R_s Cv}\right) \right] - I_{0(c)} \exp\left[\frac{\alpha_c F(U - U_{0(HER)})}{RT}\right] \quad (6)$$

where $I_{0(c)}$ is the *cathodic exchange current density* measured at $U = U_{0(HER)}$, and α_c is the cathodic charge-transfer coefficient. Equations (5) and (6) represent capacitive current plateaus ($I_c = \pm Cv$) with superimposed faradaic components. However, in the absence of water-splitting, only the first terms in these equations survive. Thus, the expected *rectangular voltammetric profiles* are verified only when $R_s \rightarrow 0$ and $I_f \rightarrow 0$, *i.e.*, current plateaus in the anodic ($I = Cv$) and cathodic ($-I = -M > Cv$) scans characterize the voltammetric response. Since in practice $\alpha_a \approx \alpha_c \approx 0.5$, it is the I_0 -values that control the extension of the current tail characteristics. Since this kinetic parameter cannot be theoretically predicted using the well-known Butler-Volmer model, the electrode material's intrinsic properties govern the practical WVV beyond the predicted thermodynamic value. Common values are in the range of *ca.* 1 to 0.1 $\mu\text{A cm}^{-2}$ for the oxygen and hydrogen evolution reactions.^[265,266] For the simultaneous *half-reactions* at the anode (+) and the cathode (-) to originate the water-splitting, it must be considered that: $\Delta E^\circ(V) = E^\circ(+) - E^\circ(-) \equiv E_{min}(V) = 1.23 + 0.0147 \log[p(\text{O}_2)/1.0 \text{ bar}] + 0.0295 \log[p(\text{H}_2)/1.0 \text{ bar}]$. Thus, one has that $U_{min} < 1.23 \text{ V}$ when the partial pressures $p(\text{H}_2)$ and $p(\text{O}_2)$ of the dissolved gases are lower than 1.0 bar. In practice, the partial pressures at the onset of the electrolysis in hermetically sealed coin cells must reside in the range of $0 < p < 1$, resulting in U_{min} -values lower than 1.23 V.^[2]

The analysis of Equations 5 and 6 shows that high ν -values promote a considerable increase in the capacitive current compared to the faradaic component. As a result, the presence of water-splitting in the VCs can be drastically masked, *i.e.*, the current is partially wasted in water-splitting, thus causing a cell precocious failure with no real gain in energy storage.

1.4. The auxiliary experiments using the EIS and SSC techniques

Despite the above considerations, low scan rates (e.g., 1 mVs^{-1}) takes a very long time to conclude the experiment and escape from the typical EDLC fast-charge regime of operation. EIS and

SSC techniques are the most powerful tools to obtain the capacitance and R_s values for EDLCs.^[267] In this sense, it is necessary to perform EIS and SSC experiments to define U_c -values and double-check the C_{EDL} and R_s values properly. Only after that, is that the *galvanostatic charge-discharge* (GCD) technique must be used to determine the specific energy and power device characteristics.

For EDLCs, the complex-plane plot (Z_{Re} vs. $-Z_{Im}$), in the absence of water-splitting, must be characterized at all voltages by a *low-frequency straight line* almost parallel to the imaginary (Z_{Im}) axis. The EIS experiments must be conducted using small cell voltage increments of *ca.* 0.1 V in relation to the open cell voltage (OCV), as in the CV experiments. The use of low-frequency conditions (e.g., $f < 10$ Hz) is mandatory in the case of the methods discussed below since only the 'low-frequency waves' can penetrate the narrow and deep pores.^[268] The *constant phase element* (CPE) approach must also be used due to the 'frequency dispersion phenomenon' inherent to all rough/porous electrodes. In light of these considerations, two different strategies can be used: (i) the '*complex-plane plot analysis*' based on the low-frequency data. Using the well-known R -CPE canonic model for EDLCs, where $Z_{CPE} = 1/Y_0(2\pi f)^n$ (e.g., $Y_0 \cong C_{EDL}$ when $n \cong 1$), good simulations are verified when $n > 0.9$ (e.g., a phase angle (φ) higher than -83°), and (ii) the '*imaginary analysis*', where only the imaginary part of the impedance data is considered. Thus, from the linearized CPE formula (e.g., $\ln(Z_{CPE}) \equiv \ln(Z_{Im}) = -Y_0 - n \times \ln(2\pi f)$) one can obtain the Y_0 and n parameters. In the latter case, only the linear region of the $\ln(Z_{Im})$ vs. $\ln(2\pi f)$ plot (e.g., $r^2 \geq 0.997$) must be considered for the linear regression's method. This method avoids the influence of the '*porous electrode behavior*' manifested at medium and high frequencies. In both cases (i) and (ii), the true (non-fractal) specific capacitance values must be extracted from the CPE using the Brug-Sluyters' formula (e.g., $C_{EDL} = Y_0^{1/n} [1/R_s]^{1-1/n}$).^[268]

In the case of SSC (single-step chronoamperometry experiments), the $I-t$ plot must be exponential for a blocked (capacitive) electrode/electrolyte interface. The SSC experiments are accomplished using small cell voltage increments (ΔU_c) of *ca.* 0.1 V, as in the case of the CV and EIS experiments. Thus, the transient current must practically disappear (e.g., $I/I_0 \leq 0.02$) at short polarization times (e.g., $t \approx 1-20$ s, depending on the electrode size). From the R -C model for EDLCs, this behavior is predicted by the $I = I_0 \times \exp(-t/\tau)$ relationship, where τ is the system's time constant ($\tau = R_s C_{EDL}$), and $I_0 = \Delta U_c / R_s$. Therefore, a current plateau after the transient characterizes the presence of a parasitic faradaic current due to electrolyte decomposition (e.g., water-splitting).

It is worth mentioning that a minimal background faradaic current in the order of μ A to mA, depending on electrode size, can be present after a transient even when the electrolyte decomposition is disregarded. Dissolved gases from the atmosphere and a very tiny amount of impurities (e.g., μ g to ng per dm³) in solutions prepared from as-purchased chemicals, which are not previously subjected to a meticulous purification process, contribute to the overall background current detected during the SSC experiments.^[269] As a result, a 'zero current' SSC

experiment is rarely achieved in practice under standard laboratory conditions.

The use of these three different electrochemical techniques (CV, EIS and SSC) permits to define unambiguously the U_c -value from the above considerations. The internal consistency between different data can be checked by comparing the C_{EDL} and R_s values determined using these techniques.

1.5. Minimum current for a EDLC prototype and the figures-of-merit

To identify the cell's maximum capacitance, one must first identify the minimum current necessary to charge it. Capacitance is a function of the applied current for charge and discharge the device so that a minimum current is related to the maximum cell capacitance. Instead of guessing an empirical value, one can use a more rational approach. In this sense, one can perform an SSC experiment at the maximum cell voltage U_c , previously estimated from the CV experiments, to identify the residual (or leakage) current accounting for the irreversible faradaic reaction. In this sense, the corresponding current value can be used to charge EDLCs. To identify that, one can wait until a *ca.* 97-98% decrease of the maximum current value (I_0), where the capacitive current practically fades away.

It can be used as a defined point in the present work the last experimental data point accessed after 97-98% of current reduction using the $\log(I)$ vs. t plot, characteristic of well-behaved EDLCs. To calculate the maximum achieved capacitance, C_{EDL} , it can be applied the following basic equation defining a "*differential capacitance*" for the special case where $C_{EDL} \neq f(U_c)$ [Eq. (7)]:^[259,270,271]

$$C_{EDL} = \frac{I_{min}}{\left(\frac{dU_c}{dt}\right)} \quad (7)$$

Thus, the *maximum energy* (E_{max}) can be calculated using the well-known equation [Eq. (8)]:^[259,270,271]

$$E_{max} = \frac{C_{EDL} U_c^2}{2} \quad (8)$$

With the U_c and R_s values on hands, the maximum power (P_{max}) is calculated. Now, one must focus on the determination of R_s . The most accurate method to determine the equivalent series resistance in EDLCs is based on the impedance technique.^[267] In this sense, using the complex-plane plot can be easily applied without using any particular model to interpret the EIS findings, *i.e.*, taking a given DC voltage corresponding to U_c , the minimum in the real impedance axis observed in the *high-frequency limit* (e.g., an ohmic resistance) is the desired R_s value (Please, see Figure 2 and Ref. [3]). Briefly, the literature presents ESR obtained from EDLCs using the direct current (DC) and alternating current (AC) methods.

Using the GCD method, the R_s value can be determined using the following relationship [Eq. (9)]:^[3,259]

$$R_s = \frac{\Delta U_{\text{drop}}}{2I} \quad (9)$$

where ΔU_{drop} is the sudden change in the voltage during the polarity reversal (*e.g.*, charge (+) to discharge (-)) and 2 is a '*rational normalization factor*' for the square-wave current perturbation function. However, the problem with this widespread method is the imprecision related to the evaluation of the ΔU_{drop} value, especially when the discharge curve is not linear.

Finally, the *maximum power* (P_{\max}) is determined as follows [Eq. (10)]:^[264]

$$P_{\max} = \frac{U_c^2}{4R_s} \quad (10)$$

The next step in this work is to perform a *numerical solution* of the theoretical model to obtain the P_L and E_L values. P_L -values range from 0 to P_{\max} according to the following equation:^[259]

$$E_L = \frac{E_{\max}}{2} \left(1 + \sqrt{1 - \frac{P_L}{P_{\max}}} \right) \quad (11)$$

Thus, varying P_L will lead to the E_L -value, where P_{\max} and E_{\max} are constants calculated as already mentioned.

It will now be put into practice an example of applying the method proposed in this work. The case study presented in the next section is based on the analysis of a standard (commercial) EDLC used to demonstrate and validate the present method, contrasting the experimental findings with the data present in the device's datasheet. It will demonstrate how to get Equations 1-3 included in this work (Please, see the Supporting Information, Section S1). As seen, a rational analysis demonstrates why the presented equations are the most appropriate ones.

2. How to construct a reliable Ragone plot: a case study using standard EDLCs

To validate this proposal, it is presented two sets of experiments. One follows the directions that were described in the previous section. The double-check analysis is performed simulating a real EDLC device as the power source to provide a load as already described.

2.1. Experimental Details

It was used in this work a standard model BCAP0005 P270 S01 EDLC from Maxwell® with the following characteristics extracted from its datasheet:^[272] Nominal mass = 2.1 g (*i.e.*, total mass of the packaged EDLC); Rated voltage (U_R) = 2.7 V; Rated

ESR_{DC} (initial) = 45 mΩ; Typical ESR_{DC} (initial) = 36 mΩ; Typical ESR_{DC} (initial at 5 s) = 70 mΩ; Rated capacitance = 5 F; Maximum stored energy (E_{\max}) = 5 mWh; Usable power = 19.3 W, and Impedance match power = 40 W. The datasheet information was double-checked by using the proposed methodology and the '*theorem of maximum power transfer*'. The latter supports a well-established methodology in electrical engineering. However, it is not easy to accomplish it in laboratory experiments or manufacturing processes to validated the electrochemical experiments. It is important to emphasize that the methodology proposed in this work is much simpler and yields the same results.

2.1.1. Applying the proposed methodology to EDLCs

The methodology discussed in the initial sections was applied to determine the *capacitance* (C_{EDL}),^[1] *working voltage window* (U_c),^[2] *equivalent series resistance* (R_s),^[3] *energy*, and *power*. That enable to construct the RP considering P_L as a function of E_L or vice-versa.

2.1.2. Applying the theorem of maximum power transfer as a double-check condition

Please see Figure 3a, b for a schematic representation of the experimental setup. Although the setup is straightforward, it is also not practical for conventional bench-scale experiments. According to the present methodology, the EDLC was charged using a model SP200 potentiostat from Biologic®. The device was then discharged using several different impedances (*e.g.*, 1 Ω, 10 Ω, 30 Ω, 34 Ω, 45 Ω, 53 Ω, 59 Ω, 65 Ω, 70 Ω, 80 Ω, 100 Ω, 108 Ω, and 500 Ω). The EDLC was charged up to 2.7 V (the maximum voltage in the datasheet), held at this value for 5 min, and discharged in the sequence using different loads. An oscilloscope was employed during the discharging process to obtain the voltage transient ($U_c(t)$) at the load terminals. Due to the low resistance of the loads, very fast reads are necessary to measure the voltage transient correctly. This procedure avoids obtaining incorrect data and misinterpretations of the discharge curve. The R_s of the device was measured using a precision ohmmeter. The electric currents in the circuit were calculated by applying Kirchhoff's law for a closed circuit ($I = U / (R_s + R_L)$) (Please, see Figure 3a).^[273] The *maximum power* on the load was calculated using the $P_L = I^2 R_L$ formula. The *power vs. time* experimental function was numerically integrated to determine the *maximum energy* [see Equation (11)].

3. Results and Discussion

3.1. Testing the proposed methodology using a commercial EDLC

At first look, Equation (11) has four variables. However, E_{\max} and P_{\max} are constants calculated using Equations (8) and (10),

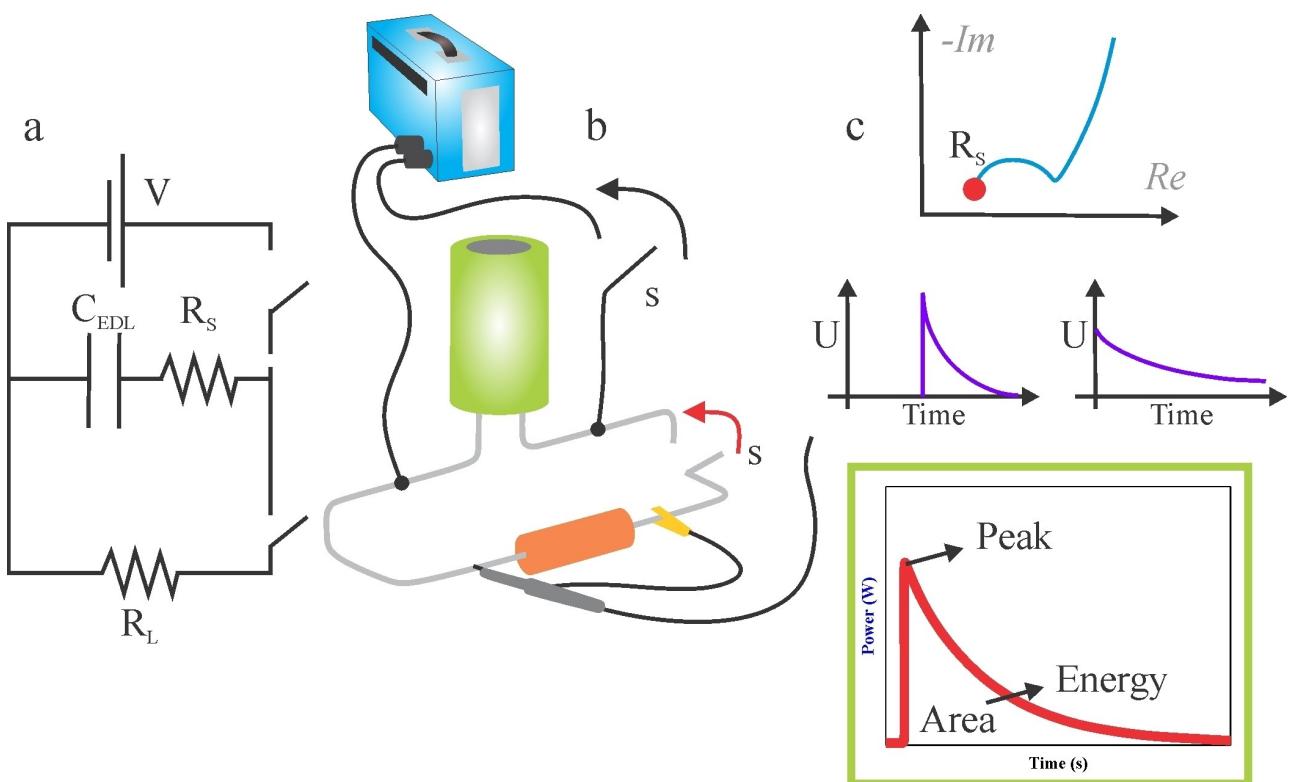


Figure 3. a) Experimental circuit containing a Maxwell's EDLC (C_{EDL}) with its internal resistance (R_s) and the variable external (load) (R_L) resistance. b) Schematic of the different circuit elements present in the setup for using a EDLC as the power source at different load resistances (R_L). c) Scheme of the different plots obtained in the experiments.

respectively. The $E_L = f(P_L)$ function is obtained numerically by assuming P_L -values in the 0 to P_{max} interval, *i.e.*, changing E_L from E_{max} to $E_{max}/2$. These boundary conditions make sense considering that for a long time discharging experiments ($t \rightarrow \infty$) the power tends to zero. In that case, one needs to identify the maximum cell voltage (U_c) and the equivalent series resistance (R_s), and calculate the maximum capacitance (C_{EDL}) of the EDLC. Then, one calculates the constants E_{max} and P_{max} and solve the Equation (11).

Figure 4 presents the experimental findings used to construct the RP. Here it is presented some hints to correctly identify the WVV using the CV (Figure 4a) and (Figure 4b) SSC techniques. The minimum current to charge the EDLC is specified in the plot. Figure 4c shows that the extrapolated high-frequency impedance data are used to determine R_s (Please, see the previous discussions in sections 1.3 and 1). With these data and considering the previous discussion in Ref. [2], it was verified a maximum cell voltage (U_c) of 2.7 V, which agrees with Maxwell's datasheet. Regarding the data in Figure 4b, the *maximum current* for charging the device was determined using $\log(I)$ vs. t plot to avoid water-splitting, *i.e.*, when the cell is held at 2.7 V after an initial current (I_0) decrease by 98%, the minimum usable current is in the 3.5–5 mA range. That value is the smallest current to ensure a non-charging process.

As a result, it is shown in Figure 4d the GCD plots registered using different currents in the specified range (*e.g.*, 3, 4, and

5 mA). The almost constant C_{EDL} -values of 4.94, 4.89, and 4.86 F determined using the Equation (7) resulted in an average capacitance of 4.90 F. These results agree with the value of 5 F informed in Maxwell's datasheet.

Using Equation (8), it was obtained *maximum energy* (E_{max}) of 4.99 mWh, in agreement with the datasheet value. To determine the *maximum power*, the next parameter to be determined is R_s . As can be seen, it was verified an R_s -value of 40 mΩ was taken from the high-frequency real impedance limit (see the inset in Figure 4c). In agreement with Maxwell's datasheet, a P_{max} -value of 45.5 W was obtained using the Equation (10).

By applying equations in Figure 1, one may obtain 2 or 3 orders of magnitude higher or lower for the energy & power values. Figure 5e contrasts different scenarios, evidencing massive distortion between the calculated values, which are far from reality.

3.2. Application of the theorem of maximum power transfer as the double-check condition for the proposed method used to construct the Ragone plot

As shown in Figure 3b, when the switch (S) is closed, the voltage on the load's terminal is a function of R_L (Please, see Figure 5a). As expected, the voltage during the charge increases as R_L is increased. By applying Kirchhoff's law,^[27] the

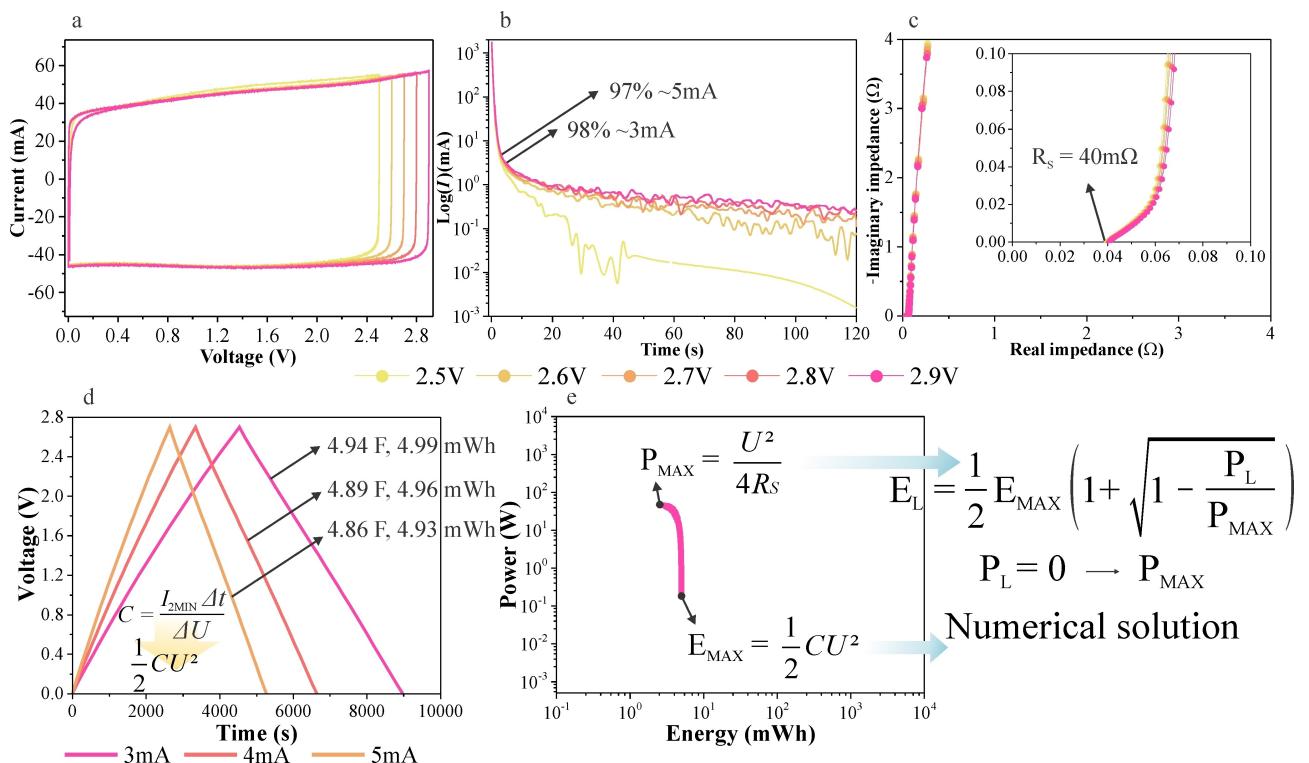


Figure 4. Electrochemical data extracted for a model BCAP0005 P270 S01 EDLC from Maxwell® highlighting: a) cyclic voltammograms at 10 mVs⁻¹ as a function of the voltage limit; b) SSC data showing the transient current in the logarithmic scale used to extract the minimum current used to charge the device; c) Complex-plane plot used to determine the R_s ; d) GCD plots obtained with the minimum current values to calculate the maximum capacitance, and e) RP constructed based on the proposed method using the numeric solution of Equation (11).

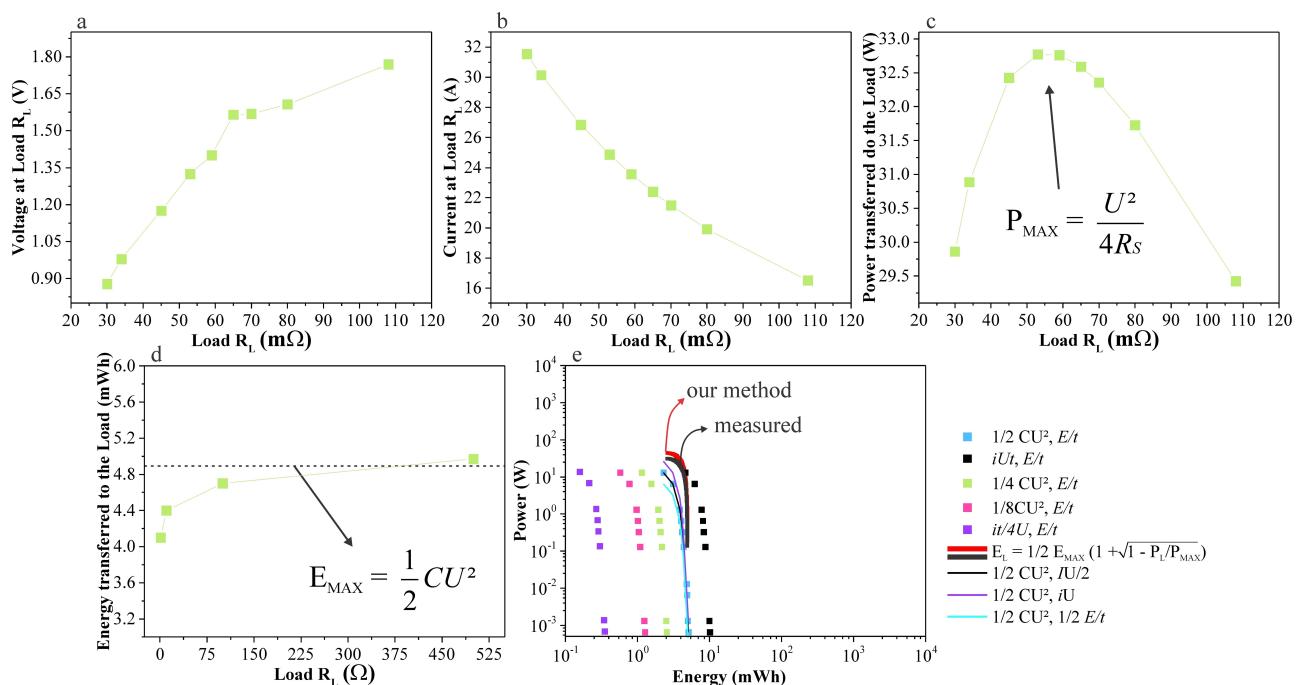


Figure 5. The electric parameters U_L (a), I_L (b), P_L (c), and E_L (d) measured in the circuit depicted in Figure 3a–b and the RP (e) as a function of the various load resistances (R_L). The RP calculated using the proposed methodology is compared with those obtained using the different equations adopted in the literature.

voltage follows the $U = (R_s + R_L)/I$ relationship. Accordingly, we verified that $I = U/(R_s + R_L)$ applying the same rules. The voltage

at the load is half of the maximum value yielding $R_s \approx 55.58 \text{ m}\Omega$. It was applied the $P_L = I^2 R_L$ equation to construct the

Figure 5c. P_{\max} is found when $R_L=R_s$, thus, the P_{\max} -values delivered by the device is 33 W, which is close to that of 40 W reported in Maxwell's datasheet. The $P-t$ data were numerically integrated to obtain the *maximum energy* as a function of R_L showed in Figure 5d. Finally, to construct the RP showed in Figure 5e, the energy & power values were combined for each resistor used in the experiments and contrasted with data presented in Figure 4e. The values obtained in this method and our calculated findings are very similar in Figure 5e.

The data used to plot Figure 5e are presented in Tables 1 and 2. As can be verified, the method proposed in this work to construct the RP (red line) agrees with the experimental data obtained for Maxwell's EDLC connected to different resistors. The black line represents the data according to the theorem of maximum power transfer. On the contrary, the use of the other equations found in the literature resulted in severe discrepancies.

4. Conclusions

It is reported in this work on how to properly construct the RPs for EDLCs. This study was motivated by the several discrepancies/anomalies verified in the literature where different equations are commonly used to obtain the energy vs. power plots. It was presented the theoretical basis of a rational method to construct the RP for EDLCs accurately. In the end, it

was verified that the main issue found in the literature regarding the RP is the '*ad hoc*' assumptions made to calculate the energy and power values. One can expect with this work to instigate a rational strategy for the new works in the field of EDLCs to standardize the methods of calculation involving the figures-of-merit used to represent the energy and power characteristics of these devices. By standardizing the methodology, as reported in this work, the use of RP becomes more useful in the field of energy storage to represent the true advances in performance obtained for newly developed devices. In this sense, when plotted in the proper manner specified in this work, the use of RP would help not just the scientific community in obtaining meaningful findings but also the manufacturers and investors to apply new materials and devices in new solutions for energy storage challenges. In this context, one expects the present work encourages different research groups to reflect on the rational use of RPs. As shown in this work, the drawbacks related to the misused of RPs can be circumvented using the rational analysis that was thoroughly discussed, which aims to contribute to new best practices inherent to the study of energy storage devices.

Supporting Information

The Supporting Information includes a table containing all the abbreviations and symbols used in the manuscript.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords: Ragone plot • Energy and power densities • Energy storage devices • Electrochemical double-layer capacitors

Table 1. Comparison of different methods used to obtain the EDLC power.					
i_c [A]	C_{EDL} [F]	Power [W] $\frac{U_c^2}{4R_s}$	iU_c [a]	$\frac{E}{2t}$	$\frac{iU_c}{2}$
0.0005	5.08	0.0007	0.0014	0.0003	0.0007
0.001	4.96	0.0014	0.0027	0.0007	0.0014
0.005	4.86	0.0068	0.0135	0.0034	0.0068
0.01	4.78	0.0135	0.0270	0.0068	0.0135
0.10	4.34	0.135	0.270	0.068	0.135
0.25	4.14	0.338	0.675	0.169	0.338
0.5	4.02	0.675	1.350	0.338	0.675
1	3.87	1.35	2.70	0.68	1.35
5	3.09	6.75	13.50	3.38	6.75
10	2.29	13.49	27.00	6.75	13.50

[a] The equations were taken from Refs. [5–258].

Table 2. Comparison of different methods used to obtain the EDLC energy.					
i_c [A]	C_{EDL} [F]	$\frac{1}{2}C_{edl}U_c^2$ [a]	Energy [mW h] $\frac{1}{4}C_{edl}U_c^2$	$\frac{1}{8}C_{edl}U_c^2$	iU_c [a]
0.0005	5.08	5.11	2.57	1.29	5.11
0.001	4.96	5.02	2.51	1.26	5.02
0.005	4.86	4.92	2.46	1.23	4.92
0.01	4.78	4.83	2.42	1.21	4.83
0.10	4.34	4.41	2.20	1.10	4.41
0.25	4.14	4.19	2.10	1.05	4.19
0.5	4.02	4.07	2.04	1.02	4.07
1	3.87	3.90	1.96	0.98	3.90
5	3.09	3.13	1.57	0.78	3.13
10	2.29	2.33	1.16	0.58	2.33

[a] The equations were taken from Refs. [5–258].

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