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The original version of this chapter was revised. The numbers in Table 1 and subsequent errors were corrected

# Modeling of Asymmetric Supercapacitor Cells Based on Electrode's Laboratorial Tests Data

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**Abstract.** This paper concerns the modeling of asymmetric supercapacitor cells based on electrode's data obtained throughout laboratorial tests, such as hydrogen bubble evolution electrodeposition. The electrode's data type here implemented concerns three electrodes system tests, which are designed to provide a single electrode output. This procedure provides instructions on the extraction of electrodes tests parameters, the device's design throughout dimensional basis and comparison with commercially available products and the estimation of energetic output by forecasting the resulting capacitance, voltage, energy and mass of the theoretical supercapacitor.

**Keywords:** Supercapacitor, Energy Storage Systems, Hybrid Supercapacitor.

## 1 Introduction

Among the various known commercial energy storage devices, supercapacitors or electrochemical capacitors, are highly rated due to their interesting characteristics such as greater power density, cyclability and longer service life [1,2], resulting in an overwhelming candidate as part of the global energy crisis solution. Such devices can be generically divided in four different types, high voltage ceramic capacitors, electric double-layer capacitors, pseudocapacitors and hybrid supercapacitors [1].

Although the mentioned characteristics, those devices lack energy density when compared to electrochemical batteries, which is fundamental when choosing a long-term device as main energy storage system for a specific implementation [1,3]. Therefore, promising materials, as is the case of electrodes, have been constantly investigated with higher hopes of surpassing the abovementioned limitation. However, it is not always possible for experimental supercapacitor cells to be produced, which can cause unwanted research setbacks or obstacles. Therefore, this document is focused on modeling of asymmetric supercapacitors of hybrid nature, composed of two different electrode materials of both capacitive and faradaic characteristics, allowing theoretical devices based on experimental electrode materials to be analyzed and implemented in energy storage systems designs, calculations and computational simulations. Furthermore, this manuscript presents the modeling procedure and calculations of an

asymmetric hybrid supercapacitor cell for widespread implementation, accounting for variances in equations regarding the laboratorial data towards cell values.

## 2 Supercapacitors for Life Improvement

Supercapacitors as a mean of energy storage are a key factor for more efficient and reliable electrical systems [1]. SC's applications encompass mobile and stationary implementations, from transportation to industrial and renewable energy requirements [4]. Macro and micro-scale applications of supercapacitors can be found in the power grid, providing support to power quality improvement that are mainly derived from the growing penetration of renewable generated power into the power grid [5]. Due to distributed generation increasing, the renewable generation can be commonly placed near consumers or even being an integrant part of consumer's electric installations. Considering this power grid point of view, the life improvement is associated with debottlenecking of transmission lines, more reliable integration of renewable energies and consequent greenhouse gas emissions reduction. Moreover, the high-power capabilities can also be addressed to mobile applications regarding power electronics as in electric vehicles (EV) [4], where it performs as a coadjutant device working in parallel with vehicle's battery energy storage system. Supercapacitors not only are able to provide extra power in EV's demand peak situations, but also contributing for an autonomy extension and batteries lifespan increasing throughout the battery cycle's reduction. The methodology addressed in following sections show the theoretical development of a generic hybrid supercapacitor based on an increased performance electrode material that results in better parameters, such as higher capacitance, energy and power density, when compared to a commercial device.

## 3 Hybrid Supercapacitor Modeling

This paper presents a hybrid supercapacitor (HSC) modeling procedure based on cyclic voltammetry (CV) and galvanostatic charge and discharge curves (GCD) data obtained for both positive and negative electrodes materials [6,7]. The approach takes into consideration the specific capacitance ( $C_{sp}$ ), potential window ( $\Delta V$ ), equivalent series resistance (ESR) and reminiscent relevant data, which were gathered from already published laboratorial experiments [6,7], assumed as primary database for the procedure development. In addition, in order to obtain the theoretical HSC physical structure, a Maxwell 2.3 V 300 F [8] pseudocapacitor was selected as device's choice of design and dimensioning intent, which original size and mass have fit the intended purpose. Therefore, the data set acquired was implemented into the developed electrode-to-cell capacitance equations, from which the individual electrodes capacitances were obtained, thus developing a correlation between electrode and cell data so it could be properly converted.

### 3.1 From Electrodes to Cell Data

In order to transpose laboratorial data into approximate real values so a device can be modeled and simulated, a correct interpretation of basic equations is required [9]. Based on the general cell capacitance and specific capacitance equations (3.1-3.2) the electrode-to-cell calculation procedure was developed.

$$C_{cell} = \frac{i \Delta t}{\Delta V} [F]. \quad (3.1)$$

$$C_{sp,cell} = \frac{I \Delta t}{m_{total} \Delta V} \left[ \frac{F}{g} \right]. \quad (3.2)$$

Where  $m$  states for the mass of active material, which correlation for asymmetric cells is defined as in (3.3),  $I$  is the current,  $\Delta t$  the time and  $\Delta V$  the voltage.

$$m_{total} = m_+ + m_- [g]. \quad (3.3)$$

In order to define the specific capacitances equations for asymmetric configurations, firstly, the correlation between cell and both electrodes capacitances must be defined. As described in [9], such correlation can be defined as a series equivalent circuit (3.4), consisting of two electrode-electrolyte interfaces represented as single capacitors  $C^+$  and  $C^-$  for the positive and negative electrodes, respectively [10],[11].

$$\frac{1}{C_{cell}} = \frac{1}{C_+} + \frac{1}{C_-} \left[ \frac{1}{F} \right]. \quad (3.4)$$

With regards to an asymmetric hybrid supercapacitor configuration, the electrode to cell capacitance is defined by the electrode with smaller capacitance (if  $C1 \gg C2$ ,  $C_{cell} \approx C2$ ) [12]. Therefore, as the greater capacitance is not predefined as either positive or negative,  $C1$  and  $C2$  are hereafter defined as  $C_{elec}^{c>}$  and  $C_{elec}^{c<}$ , respectively. Consequently,  $C_{cell}$  is defined as (3.5).

$$C_{cell} = C_{elec}^{c<} [F]. \quad (3.5)$$

Therefore, with regards to asymmetric cells, the specific capacitance can be obtained from (3.6), taking into consideration the total mass ( $m_{total}$ ) of both active materials.

$$C_{sp,asymm.cell} = \frac{C_{cell}}{m_{total}} = \frac{C_{cell}}{m_+ + m_-} = \frac{C_{elec}^{c<}}{m_+ + m_-} \left[ \frac{F}{g} \right]. \quad (3.6)$$

Based on (3.5) and (3.6) it is possible to correlate the cell's specific capacitance to the specific capacitance of  $C^{c<}$  by considering the cell's total mass as a sum of  $m^{c>}$  and  $m^{c<}$ , defined by (3.7).

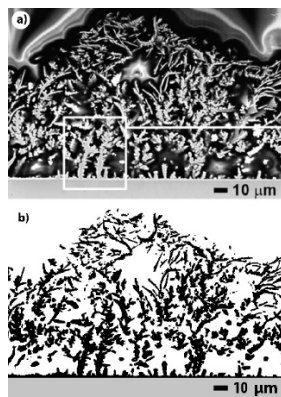
$$\frac{C_{cell}}{m_{total}} = \frac{C_{elec}^{c<}}{m^{c>} + m^{c<}} = \frac{C_{elec}^{c<}}{m^{c<}} = \frac{C_{cell}}{m^{c<}} = C_{sp,elec}^{c<} \left[ \frac{F}{g} \right]. \quad (3.7)$$

From the equations above it is possible to establish a relation between two ( $C_{cell}$ ) and three-electrodes ( $C_{elec}$ ) results for specific capacitance.

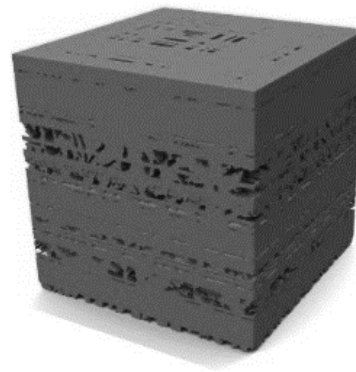
### 3.2 Electrodes Data

The nickel-copper (Ni-Cu) metallic foam developed in [6] was selected as positive electrode material for the developed HSC, whereas for negative electrode, activated

carbon (AC) [7] was implemented. The data were extracted from CVs and GCDs results, as well as scanning electron microscopy (SEM) (as shown in Fig. 1) regarding Ni-Cu electrode-to-electrolyte porosity estimation (the representation shown Fig. 2). The Ni-Cu electrode laboratorial tests [6] have presented a specific capacitance of  $105 \text{ Fg}^{-1}$  for a material deposition time of 180 s at  $1.8 \text{ Acm}^{-2}$ . From the CV results [6] a potential window of approximately 1.48 V (-0.5 V to 0.975 V) was obtained for a scan rate of  $100 \text{ mV s}^{-1}$ . With regards to the AC negative electrode, the considered specific capacitance was of  $70 \text{ Fg}^{-1}$  [7], even though [26] has presented higher laboratorial results, while a negative potential of -1.0 V with a scan rate of  $100 \text{ mVs}^{-1}$  [7, 33] was accounted.



**Fig. 1.** a) SEM  $1.8 \text{ A.cm}^{-2}$  180 s, electrode section, b) image enhancing and cleanup [1].



**Fig. 2.** Electrolyte volume from electrode porosity modelling.

Based on the SEM result addressed in Fig.1, an enhanced electrode image was obtained via various graphic software from Adobe Creative Suite 6. From the generated image, a generic section of  $100 \mu\text{m}$  was extracted and 3D modeled with the aid of the modeling software Rhinoceros 5.0 [13], which resulted in a sample of  $100 \mu\text{m}^3$  representing a proposed Ni-Cu electrode foam structure, from which the total electrode-to-electrolyte volume was obtained (and shown in Fig. 2). From the 3D model a porosity of over 89 % was noticed. Despite the difference of foams manufacturing process, when comparing the obtained result with the information gathered from [14], which stated the material's porosity in the range of 75 to 95 %, and based on the statement of [6] that metallic foams porosity values are above 50 %, it has considered that the obtained value is acceptable. Therefore, by assuming the porosity values from the manufacturer, 95 %, it was possible to calculate the respective electrolyte and electrode volumes shown in section 3.4, Table 2.

### 3.3 Generic Supercapacitor Dimensioning

The hybrid supercapacitor dimensions were based on a Maxwell 2.3 V 300 F [8] pseudocapacitor. With this purpose, a three-dimensional generic model of such device was developed determining commonly implemented thickness for each of its composing layers as well as the device's general structure. In order to individually

identify the volumetric proportion of each part of the base commercial device, a layer stack configuration was assumed based on [15], [16] and [17].

For the volume distribution supercapacitors, commonly implemented materials were applied. With regards to the device structure it was separated in the following parts: metallic case, terminals, rubber seal, electrodes, current collectors, electrolyte and separator.

As device's metallic case material, an aluminum alloy described as Aluminum 1070, with density of  $2.70 \text{ gcm}^{-3}$  and thickness of  $100 \text{ }\mu\text{m}$ , was assumed based on [18] and [19]. Regarding the device's terminal, its material was assumed as brass (CuZn37), with density of  $8.44 \text{ gcm}^{-3}$ , based on [20]. With respect to the rubber seal, the assumed material for its composition was an Ethylene Propylene Diene Monomer (EPDM), with density of  $1.40 \text{ gcm}^{-3}$  [21], [22]. With regards to the PC electrodes proposed, manganese dioxide ( $\text{MnO}_2$ ) was assumed with a thickness of  $0.5 \text{ }\mu\text{m}$  and density of  $5.02 \text{ gcm}^{-3}$  [23], [24], [25]. As current collectors an aluminum foil of  $2.70 \text{ gcm}^{-3}$  density [26] was assumed, which thickness was defined as  $30 \text{ }\mu\text{m}$  [17]. With regards to the device's electrolyte, potassium hydroxide (KOH) was assumed as it is commonly implemented as such, which density was stated at  $1.58 \text{ gcm}^{-3}$  [27]. Lastly, the separator material implemented for the developed HSC was the Celgard 2400 [28], [29], defined as a monolayer polypropylene, with thickness of  $25\mu\text{m}$  and density stated as  $0.9 \text{ gcm}^{-3}$ . Therefore, based on the presented procedure a generic pseudocapacitor configuration was defined, which resulted in a volume and mass individual distribution regarding each composing part. Furthermore, the generic device corresponded to the original mass stated in the datasheet [8],  $24 \text{ g}$ , accounting for a total occupied volume of about  $7204 \text{ mm}^3$ .

### 3.4 Ni-Cu//AC Hybrid Supercapacitor

Based on the acquired data presented in section 3.2, the Ni-Cu//AC hybrid supercapacitor was developed by replacing the electrodes materials from the generic pseudocapacitor to Ni-Cu and AC parameters, where the total volume was divided equally between positive and negative electrodes.

With regards to the developed hybrid supercapacitor, the intent was to match its final mass to the base Maxwell device. Therefore, with the new materials implementation the total mass of  $24 \text{ g}$  was obtained by optimizing the internal case volume filling by its layers stacking, which resulted in a total volume of  $14686.53 \text{ mm}^3$  (Table 2).

Therefore, the positive electrode capacitance ( $C_{elec+}$ ) was obtained by (3.8).

$$C_{elec+} = C_{sp,elec+} m_{elec+} = 638.4 [F]. \quad (3.8)$$

While the AC negative electrode, its capacitance is calculated by (3.9).

$$C_{elec-} = C_{sp,elec-} m_{elec-} = 150 \times 6.49 = 454.3 [F]. \quad (3.9)$$

**Table 2.** Ni-Cu//AC hybrid supercapacitor mass redistribution.

Item	Volume (mm <sup>3</sup> )	Mass (g)
Metallic Case	439.40	1.20
Terminals	49.80	0.42
Rubber Seal	1421.00	2.00
AC.Electrode	903.12	0.60
KOH.Electrolyte	3730.26	5.89
Current Collector (Al)	2265.34	6.12
Ni-Cu.Electrode	196.32	0.09
KOH.Electrolyte	3793.592	5.99
Separator	1887.7	1.70
Total	14686.54	24.00

Therefore, the cell capacitance ( $C_{cell}$ ) was defined by assuming the smaller capacitance value ( $C_{elec}^{c<}$ ) between both electrodes as described in section 3.1 [12] (3.10).

$$C_{cell} = C_{elec}^{c<} = 454.3 [F]. \quad (3.10)$$

With regards to the cell voltage, it was proposed based on CV data from both electrodes materials and on [30,31]. Therefore, based on the stated in [31] that the maximum charging voltage (MCV) for electrodes of different capacitances is obtained by the difference between the biggest potential value for the positive electrode (0.975 V) [6] and the smallest potential for the negative electrode (-1.0 V) [7,33], (MCV= $E_{P2} - E_{N1}$ ), the resulting proposed device's rated voltage ( $V_R$ ) is 1.975 V  $\approx$  2 V.

The equivalent series resistance (ESR) was calculated according to [32,34], which is defined by the voltage drop when the charge is interrupted, therefore, obtaining the discharge profile. In order to estimate the possible ESR value for the cell, both electrodes ESRs were obtained from the  $IR$  drop (3.11-3.12) via GCD graphic interpretation, aided by Rhinoceros 5.0 [13], therefore, the respective data was extracted for the AC electrode from [7,35,37], while for the Ni-Cu foam, from [6,36] 1.8 Acm<sup>-2</sup> at 180 s sample. The total ESR was considered as the summation of both electrode's internal resistances ( $Ri_{AC}$ ,  $Ri_{Ni-Cu}$ ) (3.13).

$$ESR_{AC} = \frac{V_{drop}}{2\Delta I} = \frac{0.07}{2 * 0.01} = 3.5 [m\Omega]. \quad (3.11)$$

$$ESR_{Ni-Cu} = \frac{V_{drop}}{2\Delta I} = \frac{0.019}{2 * 0.001} = 9.5 [m\Omega]. \quad (3.12)$$

$$ESR_{total} = Ri_{AC} + Ri_{Ni-Cu} = 3.5 + 9.5 = 13 [m\Omega]. \quad (3.13)$$

Therefore, the maximum energy storage (3.14) based on [6] and [30], gravimetric specific energy (3.15) [6] and usable specific power (3.16) [6] were calculated.

$$E_{max.hybrid} = \frac{C \cdot V_R^2}{3600} = 0.25 [Wh]. \quad (3.14)$$

$$E.sp_{hybrid} = \frac{E_{max.hybrid}}{m} = 10.42 \left[ \frac{Wh}{kg} \right]. \quad (3.15)$$



$$P \cdot sp_{hybrid} = \frac{0.12 \times V_R^2}{ESR \times m} = 1.54 \left[ \frac{kW}{kg} \right]. \quad (3.16)$$

## 4 Conclusions

The presented modeling procedure for a theoretical hybrid supercapacitor was developed in order to allow experimental promising electrode materials to be implemented in computational simulations, so their influence and interaction can be analyzed when applied to a wide range of situations, from UPSs to electric vehicle models. With regards to the procedure itself and obtained results, only publication-based analysis and verification were performed, thus experimental confirmation is defined as the next stage of this very study, characterized as future work. Therefore, when analyzing the developed Ni-Cu//AC hybrid supercapacitor results, despite the lack of experimental validation, one can propose that the obtained data has demonstrated promising values when compared to the Maxwell 2.3 V 300 F pseudocapacitor, presenting increased capacitance (454.3 F), energy (0.25 Wh) and specific energy (10.42 Wh kg<sup>-1</sup>), which makes it an interesting candidate for further study and development.

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