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Thermal and Voltage Testing and Characterization of Supercapacitors and Batteries

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Abstract - **The aim of the paper is to present and emphasize the benefits of using hybrid storage devices (composed of batteries and supercapacitors) and typical controllers in a wide range of applications, especially in the automotive field. In order to offer an optimal integration of the mentioned devices for different types of applications, the focus is oriented on an experimental study of the electrical and thermal behavior of the organic supercapacitors. The impedance spectroscopy and DC charge / discharge tests were made to observe the behavior of the fast release storage devices and to experimentally determine the electric series resistance and the capacitance variation of the supercapacitors. The experimental and simulation results are presented, analyzed and compared. Also, the mathematical model was determined by interpreting the experimental data obtained as a result of the DC charge / discharge measurements.**

I. INTRODUCTION

Lately, a great attention has been focused on energy storage devices, their most important parameters being the power and energy density. In case of the rapid release storage devices (supercapacitors) the higher is the power density, the more reduced is the necessary time for charging and discharging. In present, many researchers are trying to develop storage devices able to provide both energy and power density [1], [2].

Our main goal is to offer a hybrid device able to provide high power and energy density with a flat time response [1]. When designing such a device, there are three major problems that have to be taken into consideration. The first problem is how to choose the right devices which compose the hybrid system, the second one is related to the correct sizing of the components of the hybrid system and the third one is the inertia of these devices which limits their dynamic performance and their lifetime (especially for batteries) [3].

A modality to choose the proper devices is to identify their characteristics. Thus, we propose to study the behavior of the organic supercapacitors in order to combine them with batteries and eventually with fuel cells. There are multiple methods for electrical characterization of the storage devices but we choose two complementary methods: direct current charge / discharge (cyclic voltammetry [4]) and impedance spectroscopy [5], [6]. By identifying the electrical and thermal behavior of the internal parameters of the rapid release storage devices our hybrid device will be able to provide both power and energy density [7]. Such a device can be used in multiple applications, especially in the transportation domain because of its rapid bidirectional exchange of energy and its overall dynamic and thermal stability.

In order to connect supercapacitors and batteries in a single hybrid device, the present paper experimentally determined their behavior regarding temperature and voltage [7], [8]. The results were interpreted and the conclusions were taken.

II. BATTERIES AND SUPERCAPACITORS

Nowadays, the comfort in transportation and automotive can be improved only by increasing the power and current demands, demands which rise the overall heating of the storage devices.

In present, ***batteries*** are the most widely used energetic storage devices that can assure high energy density with reduced power density. Researchers try to improve the performances of the batteries, fuel cells and supercapacitors in order to reduce their size, weight and to increase their capacitance [4]. Also, they are focused on developing power supplies more suitable for electronic and industrial applications [6]. On the other hand, the manufacturing technology of the batteries is improving slowly compared to the requirements of the new power applications, resulting in the need of incorporating them in hybrid systems. New electrochemical models can be taken in consideration while investigating a modality to perform this integration. These models consider the electrochemical and thermodynamics processes and the physical structures when testing the discharging process of the batteries.

Accordingly to the state of the art, the power supply of the mobile devices is ensured by secondary batteries [4], [9]. Unfortunately, during the charge / discharge process of the batteries, their internal structure can be damaged due to the repeated electrochemical reactions thus reducing their lifetime. The lifetime of the battery is also influenced by external parameters such as: peak current demands, deep discharge, temperature and humidity [4]. The energy provided by a battery depends on the active materials which compose the battery itself. Also, at the battery level there is an important loss of its capacitance [5]. Practically, only a part of the capacitance of the battery can be used in the

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discharging process because its performance and capacitance are influenced by non-reactive substances, dendrites, weight, dimensions, ambient temperature and aging characteristics.

A DC charge / discharge experiment with 2 A current (illustrated in Fig. 1) demonstrates that if a 12 V / 77 Ah lead acid battery is functioning at extremes temperatures, its performances are reduced, the charging and discharging times varying with temperature [10].

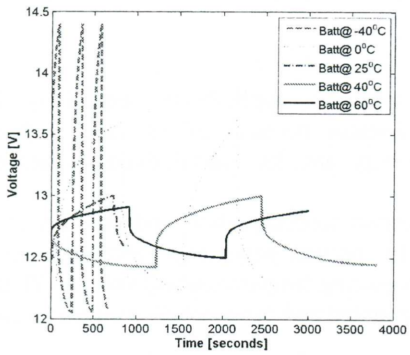


Fig. 1 Battery dependence with temperature

Instead, ***supercapacitors*** can be almost completely charged and discharged, have high power density, express more temperature stability than other energy storage devices and it is not influenced by peak currents [5].

Supercapacitors are electrochemical double layer capacitors (EDLC) characterized as being fast release storage devices able to provide very high level of power density with medium energy density. Another important aspect is related to the high cyclability of EDLC which accepts more than $0.5 \div 1 * 10^6$ cycles during its lifetime [11]. Because of their manufacturing technology and because of the improvements made in the nanotechnology field, they can offer new improved solutions for multiple applications, from automotive to biomedical and industrial ones. Their main benefits are represented by increased power density and cyclability, in comparison with the batteries that present a higher energy density. Thus, they can store a high level of power energy in a small volume and also, they can release it in power bursts, ideal case for peak power applications, such as improving the acceleration and load smoothing.

At the device level, a supercapacitor is composed of two activated carbon electrodes, a separator and an organic or inorganic electrolyte [12].

The operating principle of the supercapacitor is based on storing the energy by using the charge transfer at the interface between the activated carbon electrode and the electrolyte when applying a voltage at the terminals. The particularity of their structure (defined by the manufacturing technology) is to involve only electrostatic phenomenon to transfer the ions between the charge collectors through the porous structure [13], [14]. Their ideal operating principle is based on not including chemical reactions during charging and discharging periods (as in the batteries case) thus increasing their power density, reducing the time necessary for the charging processes and increasing their lifetime. Unfortunately, inside the supercapacitors parasitic electrochemical reactions can appear. These reactions can be more or less amplified, depending on the manufacturing technology thus reducing the lifetime of the supercapacitors [14], [15].

De Levie showed that the charging and discharging processes do not occur with the same time constant throughout the electrode material because of the finite conductance of the electrolyte which leads to a voltage drop along the pores [2], [8]. Also, inside the supercapacitor there are different kinds of pores from nano-pores to micro-pores which are not so quickly charged or discharged as the macro-pores which have smaller time constants [16].

There are two main types of supercapacitors: organic and inorganic. Lately, efforts are being made to improve the quality of both organic and inorganic supercapacitors [17]. According to the state of the art, *inorganic stacked supercapacitors* are used in "high voltage" applications, 400 V, 300-350 Wh and power range between hundreds of watts to megawatts [3], [4], [16], [18]. *Organic super-capacitors* use electrolyte material that causes limitations regarding temperature range of operation and the value of the energy density is about 2 Wh / kg [3], [4]. For transmission systems and hybrid powertrains these limitations are relevant because organic supercapacitors can be used only below 50 °C and over 20 °C in the propylene carbonate electrolyte type. Instead, aqueous supercapacitors are more stable in a wide range of temperatures. The operational voltage per cell is limited only by the breakdown potential of the electrolyte and has values less than 1 V per cell for inorganic electrolytes and less than 3 V for organic ones.

The behavior of the organic supercapacitors at different temperatures is illustrated in Fig. 6–9, 11–14.

Because of their performances, supercapacitors are suitable for applications which require high power pulses [19], [20]. Used in a hybrid battery-supercapacitor system they can extend the lifetime of the battery by taking the peak currents, can reduce the costs of the battery by resizing it, can be used in regenerative breaking and, as well, in power electronics applications such as wind turbine [21], [22].

III. HYBRID DEVICES

In the automotive field, hybrid devices are suitable for multiple applications, and especially for applications developed to improve the energy efficiency and to reduce the fuel consumption and pollution.

The ***start process*** requires a peak current which increases the temperature at the battery level thus reducing its capacitance and its lifetime. In order to eliminate this risk energetic buffers able to compensate the intrinsic limitations of the battery can be used. Supercapacitors can be considered as being such energetic buffers because they can absorb the peak currents, thus protecting the battery in the start process.

Another application used to improve the energy efficiency of the vehicles is ***regenerative breaking***, a process which becomes more efficient if the charging process of the storage device can be completed in a short time. A supercapacitor can assure this demand because of its short charging time.

Developing a hybrid system composed of batteries and supercapacitors became an important goal that need to be assessed because of its ability to provide high power and energy density. In order to observe the functionality and the advantages of the hybrid supercapacitor-battery system, the configuration illustrated in Fig. 2 was simulated. The goal of the simulations was to monitor the state of discharge of the battery in the case of a load supplied from the battery and the same load supplied from the hybrid system.

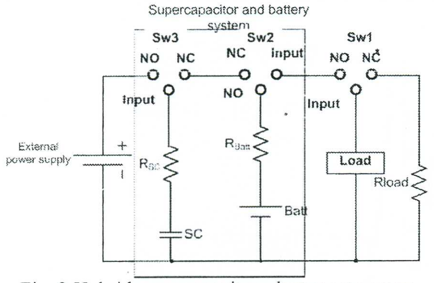


Fig. 2 Hybrid supercapacitor - battery structure

In order to make the simulations, three MOSFET transistors were used as double switches connected, as it is illustrated in Fig. 2, where NC represents the normal closed state of the switch and NO the normal opened state. Also, a 12 V / 14 F inorganic supercapacitor and a 12 V / 77 Ah lead acid battery were used as storage devices.

Thus, the initial conditions were set and the state of discharge of the battery was monitored. In order to obtain the results, 5 identical 10 seconds cycles were applied to the input of the both systems and the battery was monitored. The results of the simulations that demonstrate the advantages of the hybrid devices are illustrated in Fig. 3 and Fig. 4.

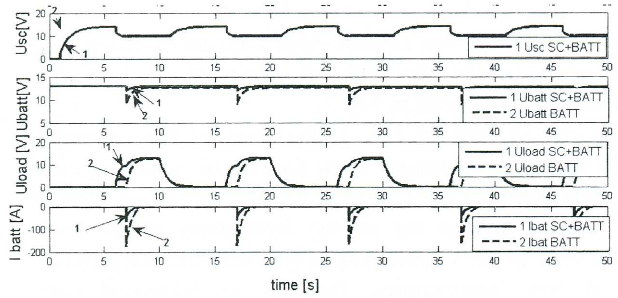


Fig. 3 Results of the simulations

As it can be seen in Fig. 3, the supercapacitor can be easily and quickly charged from an external power supply in order to provide the high peak power pulse demands of the load profile. The benefits of integrating a supercapacitor in the system described above are illustrated in Fig. 4. As it can be seen, if the battery is used as standalone storage and energy source, its state of discharge is reduced from 100 % to 99.94 % in the specified 5 cycles. Instead, if the load is supplied from a hybrid battery-supercapacitor system, the state of discharge of the battery is reduced only to 99.99%, thus the lifetime of the battery being increased. Also, the values of the currents supplied from the battery are reduced thus increasing its state of charge and lifetime.

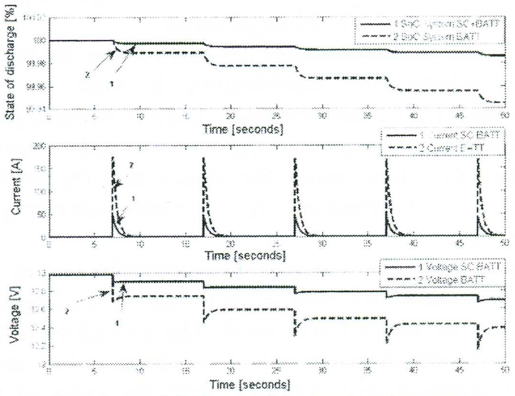


Fig. 4 State of discharge of the monitored systems

Because in simulations the supercapacitor proved to be suitable for hybrid devices, its energy performances were tested. Firstly, the value of the equivalent electric series resistance (ESR) and of the capacitance were determined, parameters which can drastically influence the amount of the energy provided by the supercapacitor. Also, its behavior in temperature and voltage was tested because these factors influence the lifetime of the supercapacitor. If the supercapacitor is functioning at high temperatures and overvoltage, the parasitic electrochemical reactions are accelerated, it appears the decomposition process of the electrolyte and thus its lifetime can be reduced [23].

In experiments, four packs of BatsCap organic supercapacitors were tested at different temperatures and in a wide range of frequencies in order to identify their behavior.

IV. EXPERIMENTS AND RESULTS

In order to determine the behavior of the storage devices it were made DC charge / discharge and electric impedance spectroscopy tests on four packs of 2700 F / 2.7 V organic supercapacitors and also, on the four packs of organic supercapacitors connected in series.

A. DC charge / discharge process

In the DC charge / discharge process on organic supercapacitors voltage balancing resistances were used. These resistances should be used when the module is composed of organic supercapacitors connected in series in order to evenly distribute the voltage across the supercapacitor. If they are not used, especially in high voltage supercapacitors, the variation in leakage current can lead to appearance of overvoltage across some supercapacitors having as a direct result gassing or explosion. This voltage difference can occur if the capacitance of a cell is lower than the ones of the other cells. The value of the balancing resistance can be determined by using (1):

$$R = \frac{T}{C \cdot \ln(\frac{V_t}{V_o})}. \qquad (1)$$

where T is the charging time until the value of the voltage on supercapacitor is rising from the initial voltage V_o to the threshold value, V_t and C is the capacitance.

The inorganic supercapacitors do not require balancing resistances because the equilibration is made in the manufacturing stage of the electrolyte. Instead, in the organic supercapacitor case, the spiral of the activated carbon cannot be connected in series inside the device and the desired capacitance can be reached only by externally connecting in series such cells.

To characterize the organic supercapacitors and to determine their ESR and their capacitance the cycling DC charge / discharge method was used. The procedure is the following: (i) the supercapacitor is charged by using balancing resistances with 5 mA / F until the value of the voltage reaches the threshold voltage of the supercapacitor; (ii) After the charging process is completed, the balancing resistances are removed and the supercapacitors are stabilized for 30 minutes; (iii) After the stabilization process is completed, the supercapacitors are discharged at 100 mA / F constant current (270 A for the 2700 F / 2.7 V organic supercapacitor) for 5 seconds. The value of the voltage after discharging at 100 mA / F for 5 seconds is stored (U_1). The 5 seconds discharging is followed by a 5 seconds relaxation of the supercapacitor and the value of the voltage is stored (U_2). U_1 and U_2 are used to determine the value of the ESR by using (2).

$$ESR = \frac{U_2 - U_1}{I}. \qquad (2)$$

In order to determine the value of the capacitance of the four packs of supercapacitors it was used (3):

$$C = \frac{I \cdot \Delta t}{\Delta U_{cap}}. \qquad (3)$$

$$\Delta U_{cap} = (0.8 \cdot U_{th} - 0.4 \cdot U_{th}) - (0.6 \cdot U_{th} - 0.3 \cdot U_{th}) \qquad (4)$$

Based on the measurements, the methodology used for sizing the hybrid battery-supercapacitor device is illustrated in Fig. 5.

A. DC measurements

To make the DC measurements at 25 °C and 50 °C for determining the values of the capacitance and of the ESR it was used the testing procedure described above, a Climate Thermal Chamber and a test bench implemented by the researchers from INRETS LTN [5], [10].

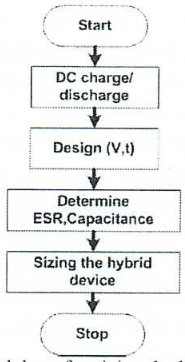


Fig. 5 Methodology for sizing the hybrid device

For the first DC experiments, the threshold voltage was set at 2.22 V. The results are illustrated in Fig. 6.

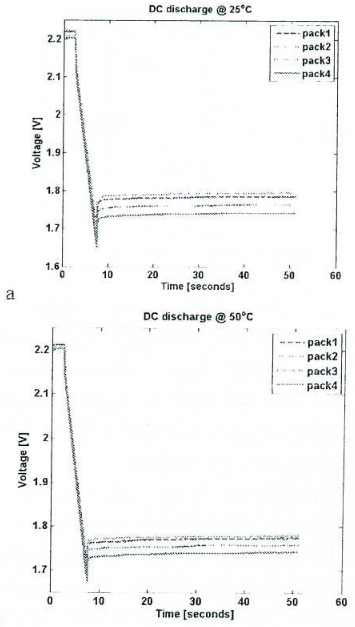


Fig. 6 DC measurement on packs of organic supercapacitors charged at 2.22 V; behavior at: a – 25 °C; b – 50 °C

The behavior of the organic supercapacitors charged at 2.22 V and heated at different temperature levels is comparatively illustrated in Fig. 7.

As it can be seen in Fig. 7, the level of the relaxation voltage was more increased when the organic supercapacitor packs were tested at 25 °C than in the case of testing them at 50 °C. This is due to the strong dependency of the ionic concentration of the organic electrolyte with the temperature. Thus, at low temperatures, there is an increased ionic concentration inside the supercapacitor, which leads to a higher level of the relaxation voltage but a reduced value of the capacitance, as it can be seen in Table II. On the other hand, at high temperatures, the ionic concentration is reduced and the capacitance is increased.

The corresponding values of ESR for the four packs of organic supercapacitors charged at 2.22 V are calculated and presented in Table I.

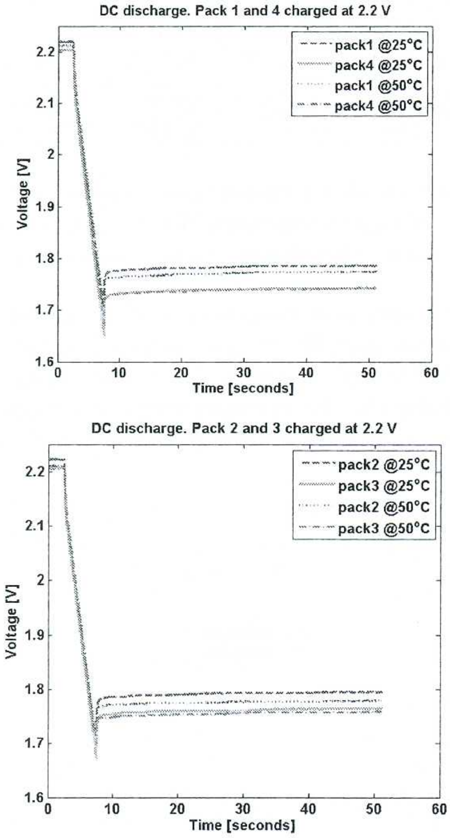


Fig. 7 Comparative behavior – packs of organic supercapacitors charged at 2.22 V and discharged with 100 mA / F at 25 °C and 50 °C; a – pack1 and pack4; b – pack2 and pack3.

TABLE I.
VALUES OF THE ESR FOR THE FOUR PACKS OF SUPERCAPACITORS CHARGED AT 2.22 V AND HEATED AT 25°C AND 50°C

T [°C]	Packs	U1 [V]	U2 [V]	I [A]	ESR [mΩ]
25°C	Pack1	1.69	1.779	270	0.33
	Pack2	1.689	1.79	270	0.374
	Pack3	1.669	1.758	270	0.33
	Pack4	1.647	1.736	270	0.33
50°C	Pack1	1.702	1.766	270	0.237
	Pack2	1.698	1.772	270	0.274
	Pack3	1.69	1.752	270	0.23
	Pack4	1.672	1.733	270	0.226

The voltages measured over the four packs of organic supercapacitors connected in series and charged at 2.22 V were:

$U_{1,25^oC} = 6.679$ V and $U_{2,25^oC} = 7.057$ V,

$U_{1,50^oC} = 6.74$ V and $U_{2,50^oC} = 7.021$ V.

Thus, $ESR_{total,25^oC} = 1.4$ mΩ and

$ESR_{total,50^oC} = 1.04$ mΩ.

As it can be seen, the value of the ESR corresponding to the four packs of supercapacitors connected in series is equal to the sum of the four independent determined ESR values.

Also, for the four packs of organic supercapacitors charged at 2.22 V the values of the capacitance from the acquired data by using (3) and (4) were calculated. The corresponding values are presented in Table II.

TABLE II.
VALUES OF THE CAPACITANCE FOR THE FOUR PACKS OF SUPERCAPACITORS CHARGED AT 2.22 V AND HEATEAD AT 25°C AND 50°C

T [°C]	Pack1 - C [F]	Pack2 - C [F]	Pack3 - C [F]	Pack4 - C [F]
25°C	2598.2109	2627.1028	2606.5863	2570.1614
50°C	2880.392	3040.936	2978.641	2600

As it can be seen, the four packs of organic supercapacitors are dependent of temperature, their capacitance values increasing with temperature.

Another experiment in which the threshold voltage is set at 2.72 V is illustrated in Fig. 8.

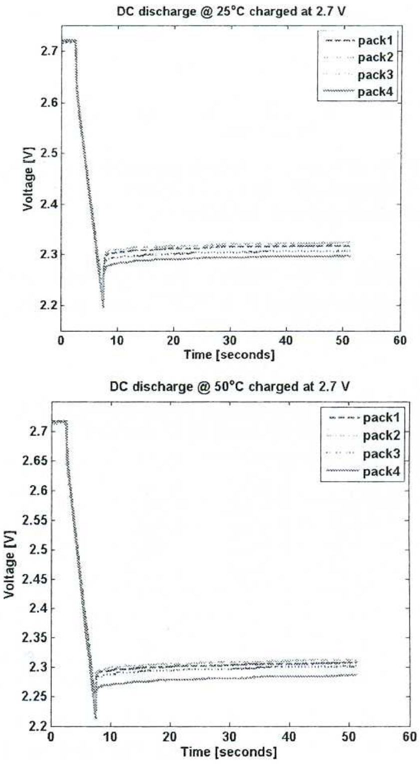
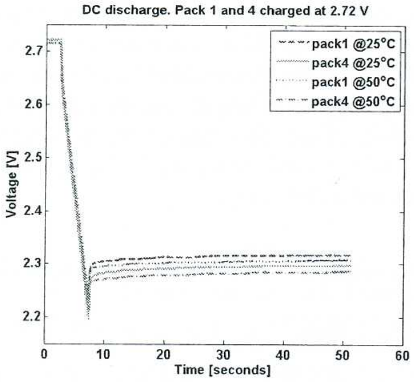
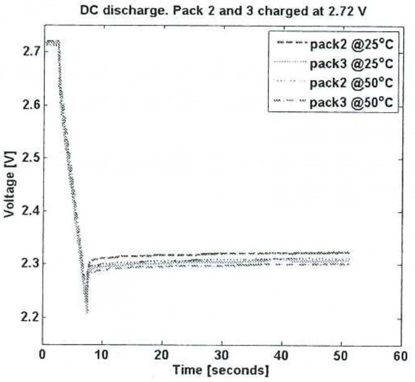


Fig. 8 DC measurement on packs of organic supercapacitors charged at 2.72 V and heated at 25 °C and 50 °C

The characteristics of the organic supercapacitors at different temperature levels and charged at 2.72 V are comparatively illustrated in Fig. 9. As it can be seen, the voltage level is more increased when the organic supercapacitor packs are working at 25°C than in the case of working at 50 °C.



a



b

Fig. 9 Comparative behavior – packs of organic supercapacitors charged at 2.72 V and discharged with 100 mA / F at 25 °C and 50 °C
a –pack1 and pack4; b –pack2 and pack3.

The corresponding ESR values for the four packs of organic supercapacitors charged at 2.72 V are calculated and presented in Table III.

TABLE III.
VALUES OF THE ESR FOR THE FOUR PACKS OF SUPERCAPACITORS CHARGED AT 2.72 V AND HEATEAD AT 25°C AND 50°C

T [°C]	Packs	U1 [V]	U2 [V]	I [A]	ESR [mΩ]
25°C	Pack1	2.212	2.306	270	0.348
	Pack2	2.211	2.315	270	0.385
	Pack3	2.208	2.297	270	0.33
	Pack4	2.193	2.285	270	0.341
50°C	Pack1	2.23	2.297	270	0.248
	Pack2	2.227	2.303	270	0.281
	Pack3	2.227	2.292	270	0.241
	Pack4	2.212	2.273	270	0.226

The values of the ESR of the packs of organic supercapacitors connected in series and charged at 2.72 V were:

$ESR_{total,25^oC} = 1.4925$ mΩ and

$ESR_{total,50^oC} = 1.085$ mΩ.

As it can be seen, the value of the ESR corresponding to the four packs of supercapacitors connected in series is equal to the sum of the four independent determined ESR values.

Also, for the four packs of organic supercapacitors charged at 2.72 V the values of the capacitance from the acquired data by using the formula (3) and (4) were calculated. The corresponding values are presented in Table IV.

TABLE IV.
VALUES OF THE CAPACITANCE FOR THE FOUR PACKS OF SUPERCAPACITORS CHARGED AT 2.72 V AND HEATEAD AT 25°C AND 50°C

T [°C]	Pack1 - C [F]	Pack2 - C [F]	Pack3 - C [F]	Pack4 - C [F]
25°C	2630.648	2549.02	2914.23	2584.795
50°C	3116.803	3250	3150.713	3071.283

As in the previous case (supercapacitors charged at 2.2 V), the four packs of organic supercapacitors are dependent with temperature, their capacitance values increasing with temperature.

The measurements were made in order to characterize the packs of supercapacitors. For the first pack of supercapacitors charged at 2.72 V and heated at 25°C, the experimental results were interpreted, the corresponding mathematical and electric model was obtained and it is illustrated in Fig. 10.

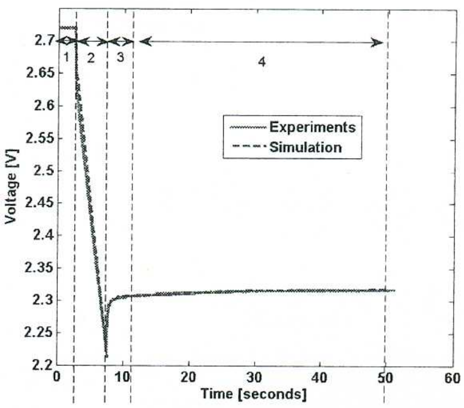


Fig. 10 DC charge/discharge experiments vs. simulation

As it can be seen, the graph from the experiments can be divided in four regions "1", "2", "3" and "4". The simulated data closely match the experimental ones.

B. *Impedance Spectroscopy method*

The second method used to characterize the supercapacitors is impedance spectroscopy method which consists in exciting the supercapacitor with a small alternative (voltage / current) signal and monitoring the current / voltage at the output stage [4], [9]. This method is used to characterize the dependency of the supercapacitor with frequency and to determine the values of the internal parameters of the supercapacitor, such as: ESR, capacitance, total impedance, the dependency between the real part and the imaginary part of the measured impedance.

For impedance spectroscopy measurements a Climate Thermal Chamber that stabilizes the temperature inside the supercapacitor and a Zahner IM6 impedance analyzer were used. The IM6 device automatically extracts the real and imaginary part of the measured impedance at different frequencies. As range of frequencies, 31 points of frequencies were chosen. The IM6 device divides the frequency range in high frequency range (f > 66 Hz) and low frequency range (f < 66 Hz). In the measurements it was chosen the 10 mHz - 1 kHz frequency range which corresponds to the typical time constants in the most high power applications [5]. To reduce the errors that appear because of the coupling effects between current and voltage twisted pairs were used.

The measurements made on the four packs of supercapacitors heated at 0 °C, 25 °C, 50 °C are illustrated in Fig. 11, Fig. 12, Fig. 13 and Fig. 14.

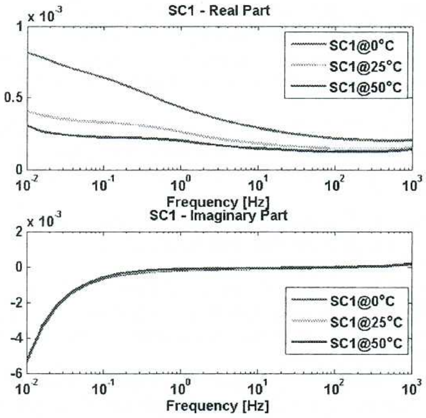


Fig. 11 Impedance spectroscopy on "SC1" organic supercapacitor pack1

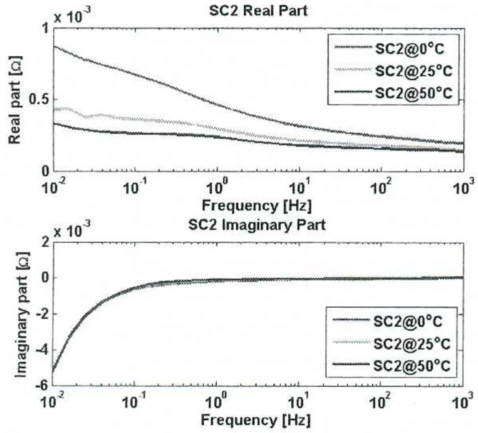


Fig. 12 Impedance spectroscopy on "SC2" organic supercapacitor pack2

The graphs are illustrated in logarithmic scale. As it can be seen in Fig. 11 - Fig. 14, the behavior of the parameters of the packs of organic supercapacitors is influenced by the frequency variation. Thus, at low frequencies the real part of the measured impedance has increased values and at high frequencies the values of the real part of the measured impedances are reduced. Also, the absolute value of the imaginary part of the measured impedance decreases until the value of the frequency reaches 1 Hz and after that, the values of the imaginary part of the measured impedance is maintained approximately constant.

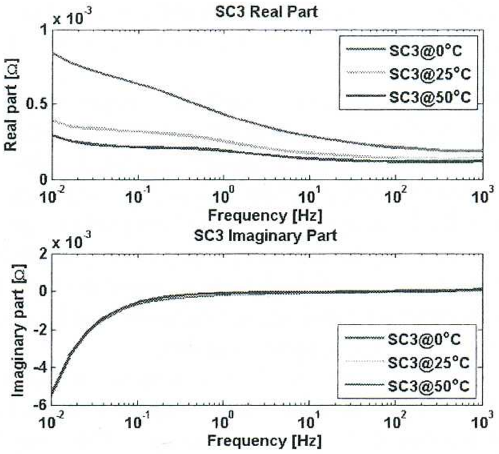


Fig. 13 Impedance spectroscopy on "SC3" organic supercapacitor pack3

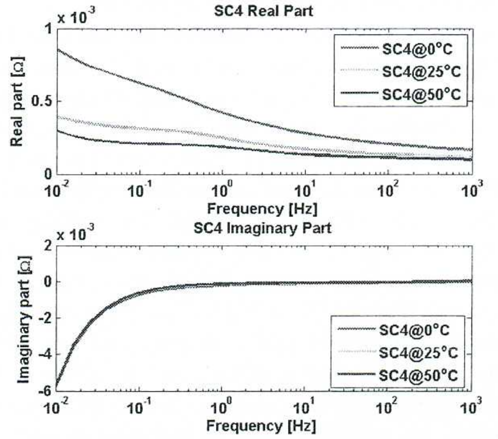


Fig. 14 Impedance spectroscopy on "SC4" organic supercapacitor pack4

As it can be seen, the real part of the organic super-capacitors varies with almost 0.6 mΩ (for 0 °C and 50 °C), value that can be neglected in automotive applications. The variation of the imaginary part with temperature is almost insignificant, as it can be seen in Fig. 11 - Fig. 14. Thus, even if organic supercapacitors are dependent with temperature, because the variation of the real and imaginary part is not increased, they can be considered as being a real alternative for the actual storage devices used in transportation field.

V. CONCLUSIONS AND FUTURE WORK

To observe the behavior of two types of devices connected together in a hybrid battery-supercapacitor device, simulations were made. The results proved that the hybrid system is suitable for a wide range of applications. For sizing the hybrid device, the ensemble had to be characterized. As result, two complementary methods were used for characterizing the organic supercapacitors: DC charge / discharge and impedance spectroscopy.

The cyclic DC charging / discharging method was used to determine the values of the ESR and of the capacitance for the four packs of organic supercapacitors. Also, the experiments were relevant from the point of view of the device characterization.

Based on the DC charge / discharge experiments on BatScap 2600 F / 2.7 V organic supercapacitors, a model was developed. The corresponding model endowed with the experimental coefficients certified that our methodology for organic electrolyte supercapacitor characterization can be used in order to improve the design process of the hybrid energy sources. The methodology brings a more detailed data related to the temperature and voltage behavior that facilitates the sizing process of mobile power supplies.

Impedance spectroscopy method was used in order to determine the variation of the parameters of the four packs of organic supercapacitors with frequency. This method revealed the importance of temperature variation for these kinds of supercapacitors, especially around and below zero centigrade, fact that confirms the literature on this topic.

Supercapacitors proved to be suitable for multiple high peak power applications and they can be used in a wide range of temperature, without having an important change in their performances. Practically, the variation of the capacitance and of the ESR of the organic supercapacitors can be neglected if they are used in usual automotive applications.

The future work will be focused on testing and characterization of temperature and voltage behavior of the stacked aqueous supercapacitors (which can extend the temperature range, from -40°C to 70÷80 °C). The possibilities to embed control functions and strategies that can drive optimally the power flow to actuators and to obtain a flat response over time of hybrid devices according with specific applications will be analyzed. Also, a real time system for estimating the parameters of the hybrid device will be implemented.

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