

Experimental study of supercapacitor serial resistance and capacitance variations with temperature

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Abstract

This paper treats an experimental study of the electrical and the thermal behavior of supercapacitors for power electronics and transportation applications. In this study, the charge and discharge of supercapacitors have been characterized by taking into account the temperature variations of the device and its environment. Thermal dependence of supercapacitor serial resistance and capacitance is determined experimentally. An equivalent circuit is proposed to describe the electrical and the thermal behavior of the supercapacitors. The equivalent circuit is implemented in Saber and Spice software for simulations. Experimental and simulation results are presented, analyzed and compared. © 2003 Elsevier Science B.V. All rights reserved.

Keywords: Supercapacitor; Electric energy storage; Thermal behavior of supercapacitor

1. Introduction

In last years, a great attention has been focused on double-layer capacitors or supercapacitors in the United States, Europe and Japan. Much of this work is directed towards transportation. The first supercapacitor development program was initiated in 1989. Today several companies, such as Maxwell, Siemens Matsushita (EPCOS), NEC, Tokin commercialize supercapacitors. Potential applications concern short time uninterrupted power supplies and peak load in combination with batteries or fuel cell [1–3]. The power density of supercapacitors is considerably higher than that of batteries, and the energy density is higher than that of electrolytic capacitors for power applications. Supercapacitors store high level of energy in a small volume and then release this energy in power burst.

Because supercapacitors move electrical charges between conducting materials, rather than perform any chemistry, they maintain a cycle ability far longer than batteries.

Supercapacitors can be used in numerous applications, in electric or hybrid vehicles in order to provide peak power for improved acceleration, for energy recovery, in parallel with the vehicle battery during start up of a thermal engine with the purpose of decreasing the size and the power of the

battery, in fuel cell vehicles in order to reduce the power and therefore the cost of the fuel cell.

Efficiency is also a very important issue for supercapacitor in electric or hybrid vehicles applications. Part of the available energy is dissipated at the internal resistance, effective series resistance (ESR). At high power and high current, this losses can become dominant. In a recent comparison of supercapacitor and batteries in electrical vehicle applications, Burke and Miller [4] and Conway and Pell [5] found that there is a slight advantage of a good capacitor over a good battery in terms of round trip efficiency, the efficiency of the capacitor being 92% and that of a NiMH battery about 85%.

All electrochemical energy storage devices experience changes of temperature upon charging and discharging. In the case of batteries and fuel cells, the thermal effects are of from the joule heating and from the heat change generated from the Faradic cell reaction. In the supercapacitor there is no Faradic processes.

Supercapacitor performances and life cycle depend on temperature. However, electrolyte conductivity is one of the most important properties that is temperature dependent. So, thermal variations of the electrolyte ionic conductivity and activated carbon conductivity determine the thermal law evolution of ESR and of the capacitance.

The equivalent serial resistance is important during charging and discharging, because of the power dissipation that will cause internal heating, especially in power electronic applications. For this reason, it is necessary to study the ESR

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thermal variations in order to predict the power losses in the system.

To use supercapacitors in electric or hybrid vehicles, we need to know their electrical and thermal behavior, in order to simulate, design and optimize the system. For this reason, we present in this paper the electrical and thermal behavior of 2700 and 3700 F supercapacitors. Presentation and description of operating supercapacitors are presented. An equivalent electrical circuit by taking into account thermal variations of the device and its environment is proposed to describe the supercapacitor electric and thermal behavior in transportation applications. Determination of the equivalent electric circuit from experimental results is discussed and analyzed. The model has been implemented in Saber and Spice software. Finally, experimental and simulation results are compared.

2. Supercapacitor description

The unit cell of a supercapacitor is based on the double-layer capacitance. The elementary structure of supercapacitor consists of two activated carbon electrodes and a separator that prevents physical contact of the electrodes but allows ion transfer between them.

Energy is stored in the double-layer capacitor as a charge separation in the double-layer formed at the interface between the solid electrode material surface and the liquid electrolyte in the micropores of the electrodes.

When a dc voltage is applied as shown in Fig. 1, the electric double-layer is formed to store electric energy [3]. The double-layer capacitance is proportional to the surface area of the electrode and inversely proportional to the thickness of the double-layer. This thickness is in the order of few angstroms. The supercapacitor capacitance is given by

$$\left(\frac{1}{C_{dl(1)}} + \frac{1}{C_{dl(2)}} \right)^{-1}$$

where $C_{dl(1)}$ and $C_{dl(2)}$ are the electric double-layer capacitance at the two electrodes.

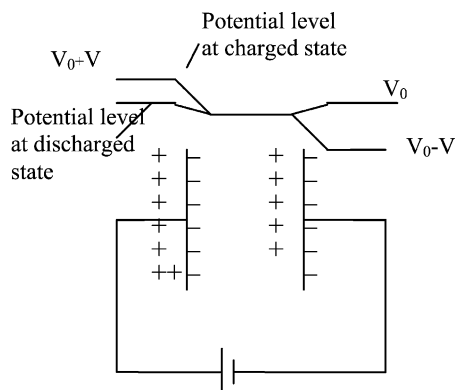


Fig. 1. Principle of operation of a single supercapacitor cell.

Supercapacitor is a device that consists of a pair of ideally polarizable electrodes; only devices that do not exhibit Faradic reaction over the potential range of operation are considered as electrical double-layer capacitors, and all the charges accumulated are used to build-up a double-layer between the conductor and the electrolyte. Charge and discharge of supercapacitor is, in fact, charge and discharge of the double-layer, because no electrochemical reaction is involved.

The ions, displaced in forming the double-layers in the pores are transferred between the electrodes by diffusion through the electrolyte [6–8]. The quantity of energy and charge stored in supercapacitors are a function of the supercapacitor capacitance and its serial resistance [9–11]. Moreover, to maximize the capacitance in order to store the maximum of energy, the activated carbon surface area must be maximized and the double-layer thickness minimized.

The science and technology of supercapacitors are reviewed for a number of electrode materials, including carbon, mixed metal oxides, and conducting polymer. Activated carbon is the electrode material used most frequently for supercapacitors. The reasons are low cost, high surface area, availability, and easy production technology. Activated carbon is available with a specific surface area of up to 2000 m²/g; this corresponds to a theoretical value of the specific capacitance of up to 500 F/g. In practice the supercapacitor specific capacitance is lower, and it is affected when an organic electrolyte is used. Burke [1] provides a comparative table of the specific capacitance of different electrode and electrolyte materials. For example for hydrous RuO₂ electrode material and H₂SO₄ electrolyte, the specific capacitance is 650 and 100 F/g with carbon cloth electrode material and organic electrolyte. Moreover, the performance characteristics of supercapacitor depend on the properties of the electrolyte. Two solutions are proposed. The first one is aqueous electrolyte. It limits the unit cell supercapacitor voltage to typically 1.5 V, thus reducing the available energy significantly. Advantages of the aqueous electrolyte are higher conductivity (0.8 S/cm for H₂SO₄) and low cost. The second solution uses organic electrolyte, which has the advantage of a higher achievable voltage, typically 2.5 V. The cell voltage is most probably limited by the water content in the electrolyte. The organic electrolyte has a significantly higher specific resistance (the conductivity is typically 0.05 S/cm). This last also affects the equivalent distributed resistance of the porous layer and consequently reduces the maximum usable power, which depends on the total ESR of the supercapacitor. However, part of the reduction in power is compensated by the higher cell voltage achievable with an organic electrolyte. In our study, we have characterized supercapacitors, which use activated carbon electrodes and organic electrolyte.

It is well known that the power output capability of electrical capacitors depends strongly on the series resistance, which needs to be minimized [4].

There are at least four different contributions to the ESR originating from the

- electrolyte including separator;
- current collector;
- porous layer including contact to current collector;
- other contact resistances.

In order to optimize the supercapacitor serial resistance one must choose well all the materials. Another parameter, which is important for the limitation of the supercapacitor power output, is the temperature. The high electrolyte resistance also affects the equivalent distributed resistance of the porous layer and consequently reduces the maximum usable power.

3. Determination of supercapacitor model and parameters

To use supercapacitor in transportation applications, it is necessary to study its electrical and thermal behavior in its operational environment. Moreover, we need to establish an electric model of supercapacitor for simulation purposes, which takes into account temperature variations. The final aim is to optimize the operation of the system including supercapacitor.

The electrical model of supercapacitor includes

- a specific capacitance which should be defined as double-layer capacitance per gram of activated carbon (F/g);
- a serial resistance (ESR) which is a function of the electrolyte and electrodes conductivities and the contact resistance;
- a parallel resistance, which represents leakage effect.

These parameters can be determined by Gouy and Chapman theories [12,13]. In fact, practical situations are more complicated. Measured capacitance of activated carbon shows a non linear relationship with their surface area because of the types activated carbon used and their treatments. To establish an equivalent electric circuit for supercapacitor who takes into account these problems, Conway [14], and other authors [3,15–17], propose an equivalent circuit based on transmission-line model, which involves distributed capacitance C_i and resistance R_i , R_i and C_i can be considered as resistance and capacitance of the pores with certain pore size. However, in power electronic applications, the supercapacitor electric behavior can be described by an equivalent electric circuit with two RC branches [18,19]. This model is not complex and the simulation time is reduced compared with the model of transmission line.

Fig. 2 shows the equivalent circuit of supercapacitor used in this study. The purpose of this equivalent circuit is to provide a model of the terminal behavior of the double-layer capacitor in power electronics circuits. Therefore, the fol-

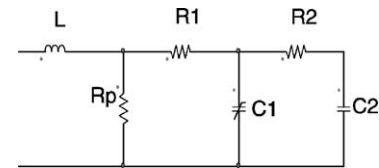


Fig. 2. Equivalent circuit of supercapacitor.

lowing requirements have been set before formulating the equivalent circuit structure:

- The model should be as simple as possible and should describe the terminal behavior of the supercapacitor over range of a few minutes with sufficient accuracy.
- The parameters of the proposed model should be determined using measurements at the supercapacitor terminals.

To decide the structure of the equivalent circuit, three major aspects of the physics of the double-layer capacitor should be taken into account [18]. First, based on the electrochemistry of the interface between two materials in different phases, the double-layer capacitance is modeled by a two parallel resistive capacitive branches with different time constants. Second, based on the theory of the interfacial tension in the double-layer, it can be expected that the capacitance of the device depends on the potential difference. In the practical range of the double-layer capacitor, Zubieta and Bonert [18] have obtained that the differential capacitance measured experimentally varies linearly with the capacitor voltage. Third, the double-layer capacitor shows certain self-discharge. A series inductor may be added for pulse applications, but measurements showed that the inductance is so small (nano Henrys (nH)) that it can be neglected in most applications.

Based on the desire for a simple model and the experience from measurements, a model of two branches is proposed. This provides two different time constants to model the different charge transfers, which provides sufficient accuracy to describe the terminal behavior of the supercapacitor.

To reflect the voltage dependence of the capacitance, the first branch is modeled as a voltage dependent differential capacitor C_1 . It consists of a constant capacitor and a capacitor whose value varies linearly with the voltage V_1 ; $C_1 = C_0 + C_v V_1$. R_1 is the equivalent serial resistance. The $R_1 C_1$ branch dominates the immediate behavior of the supercapacitor in the time range of seconds. The $R_1 C_1$ cell is the main branch, which determines energy evolution during charge and discharge cycles in power electronics applications (charge and discharge in a few seconds). It is called a fast branch. The $R_2 C_2$ cell is the slow branch; it completes the first cell in long time range in order of a few minutes and describes the internal energy distribution at the end of the charge (or discharge). R_p is the equivalent parallel resistance. The later has only impact on long term storage performances since it is a leakage effect. R_p can be neglected

during fast charge/discharge of the supercapacitor. A series inductance may be added for pulse applications, but measurements show that it is so small (some nH) that it can be neglected in power electronic applications.

In this study, we propose an experimental method to determine the ESR and the capacitance variation with temperature.

The parameters of the proposed model with two RC branches, can be identified carrying out a single fast current controlled charge. The parameters are identified by charging the supercapacitor, at different temperature values, from zero to rated voltage and by observing the terminal voltage during the internal charge redistribution (Fig. 3), the current of charge being constant (I_{ch}).

The approach to determine the different parameters is based on the fact that the two equivalent branches have distinctly different time constants. Therefore, the transient process of each branch can be observed independently by measuring the terminal voltage as function of time. It assumed that the response to the fast controlled charging process is determined only by the parameters of the fast or immediate branch R_1C_1 . After the external charging stops, all charge is in the capacitors of the immediate branch. Then, the charge redistributes itself to the second branch R_2C_2 . Zubieta and Bonert [18] have established an experimental method to determine the parameters of the supercapacitor equivalent circuit.

When the current source is switched on, the current rises to the set value I_{ch} in less than 20 ms. After this time, the supercapacitor voltage is mainly determined by the voltage drop at ($R_1 = \Delta V/I_{ch}$).

At the beginning of supercapacitor charge C_1 is determined by

$$C_0 = \frac{1}{dV/dt(0)}$$

The capacitor C_v which value varies linearly with the voltage is obtained by when $I_{ch} = 0$. Total charge supplied to the

double-layer capacitor is proportional to I_{ch} and Δt (Fig. 3). C_v is determined by

$$C_v = \frac{2}{\Delta V_t^2} (I \Delta t - C_0 \Delta V_t)$$

At the end of charge, the charge redistribution from the immediate branch to the second branch takes place. R_2 and C_2 can be determined by [18]

$$C_2 = \frac{Q_t}{V_3} - \left(C_0 - \frac{1}{2} C_v V_3 \right); \quad R_2 = \frac{V_f - (\Delta V/2)}{C_1} \frac{\Delta t}{\Delta V}$$

4. Experimental results

4.1. Experimental setup

The experimental setup consists of a supercapacitor charger based on an unidirectional current serial chopper composed of a MOSFET power switch and a fast recovery rectifier diode used as a free wheel diode necessary to evacuate accumulated energy in the high power inductance. The supercapacitors are put inside a temperature controlled climatic room. The temperature can vary between -40 and 50°C .

A data acquisition system is used to process the output thermocouple signals and the different current and voltage sensors. The acquisition system is controlled through the LABVIEW Software.

In this study, we have used 2700 and 3700 F supercapacitors with activated carbon electrodes and organic electrolyte. As this method is based on experimental results to determine the parameters of the supercapacitor equivalent circuit, it can be generalized to different supercapacitor technologies.

Experimentally, we showed that the variation of the slow branch R_2C_2 according to the temperature is very small. Consequently, in dynamic regime we can neglect this thermal variation of R_2C_2 . From the experimental results, it can

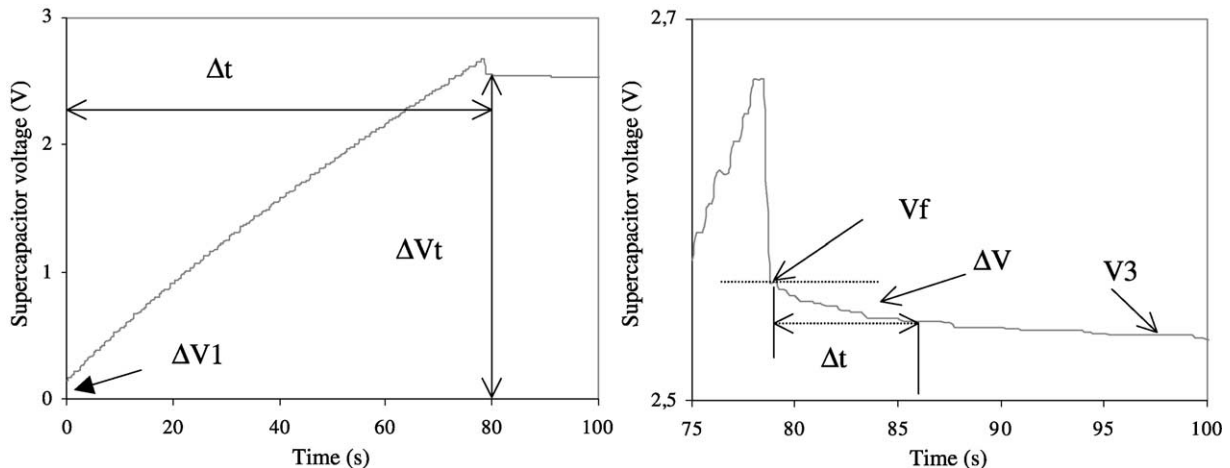


Fig. 3. Equivalent model parameters determination.

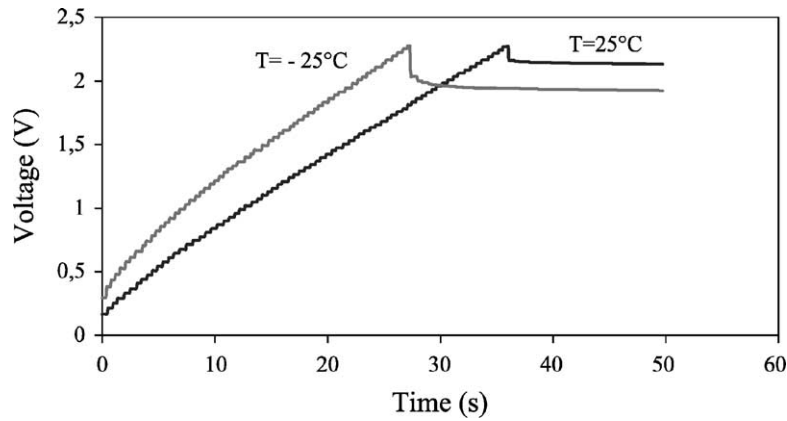


Fig. 4. Evolution of 2700F supercapacitor charge cycle.

be deduced the parameters R_2 and C_2 used in the equivalent circuit through the method described in Section 2. R_2 and C_2 are in order of 1Ω and 150 F for 2700 F , respectively.

The voltage variations across the 2700 F supercapacitor are given for two values of temperature in Fig. 4. A constant charge current of 140 A is used. The experimental curves represented in Fig. 4 show that ΔV decrease when the temperature increases. Hence, we can observe that ΔV variation at -25°C is greater than the ΔV change at 25°C .

4.2. ESR variation versus temperature

Analytical determination concerning the serial resistance variation as a function of temperature is very complex. This is due to the fact that it is very difficult to know the exact dimensions of the double-layer thickness and section. Moreover temperature dependence of electrical and chemical characteristics of the different constituents composing the supercapacitor is not known. Hence, we therefore propose to establish an empirical law using experimental results. The supercapacitor charge and discharge process have been recorded for different temperature values in a climatic room. Serial resistance has been determined using the method de-

scribed in Section 3. Fig. 5 gives R_1 variations as a function of temperature. These results show that the resistance R_1 varies greatly for negative temperature values, but low variations are measured for positive temperatures. For example, we have measured for R_1 , approximately $2.13 \text{ m}\Omega$ at $T = -25^\circ\text{C}$ and $1.1 \text{ m}\Omega$ at 25°C . It is thus clear that serial resistance decrease as the temperature increases. When the temperature rises from -25 to 25°C , the resistance varies by about 50%. This effect thus influences power dissipation by Joule effect. Consequently, for negative temperatures, the power dissipation is higher than for positive temperatures, and thus, the supercapacitor power efficiency decreases.

In order to correlate serial resistance and temperature, we have approximated the experimental values by a quadratic equation. We have thus set up an empirical law

$$R_1(T) = aT^3 + bT^2 + cT + e,$$

where a – c and e are experimentally determined constants.

For a 2700 F Maxwell supercapacitor, $a = -1.10^{-5} \text{ m}\Omega (\text{C}^\circ)^{-3}$, $b = 7.10^{-4} \text{ m}\Omega (\text{C}^\circ)^{-2}$, $c = -205.10^{-4} \text{ m}\Omega (\text{C}^\circ)^{-1}$ and $e = 1.19 \text{ m}\Omega$ the difference between measured values and the empirical law is less than 4%.

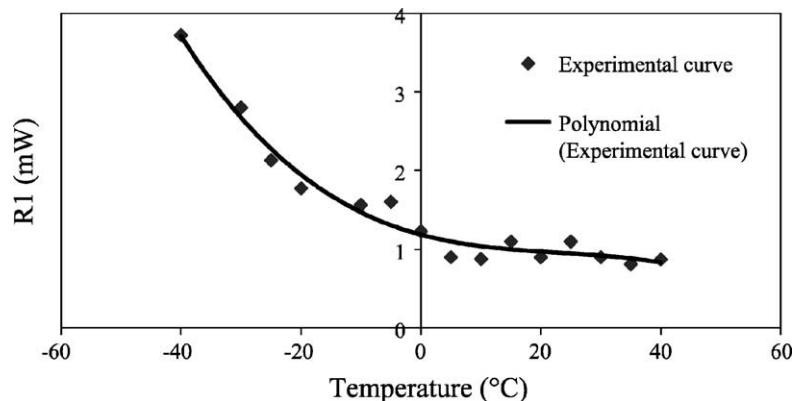


Fig. 5. Evolution of 2700F serial resistance (R_1) as a function of temperature.

4.3. Thermal variation of supercapacitor capacitance with temperature

In Section 3, we have shown that a supercapacitor cannot be modeled by a simple capacitor. In order to take into account the different physical phenomena taking place during the functioning of the device, two RC cells must be used in the model. For this reason, it is quite difficult to determine an analytic relation between capacitor and temperature. Moreover, the supercapacitor global capacitance depends on the electrolyte dielectric constant and on the thickness of the double-layer [1,14]. The variations of these parameters with temperature lead to a variation of the supercapacitor capacitance behavior as a function of temperature.

Fig. 4 shows that the charging time duration is larger at $T = 25^\circ\text{C}$; the time duration is in order of 36 s; that at $T = -25^\circ\text{C}$; the time duration of charge is in order of 27 s. This difference is due to two different effects. The first one is due to temperature variation of the supercapacitor serial resistance, and consequently, temperature variation of the time constant. Indeed, the voltage across the supercapacitor can be described by the following equation: $V(t) = R_1 I_{\text{ch}} + V_c(t)$, where $V_c(t)$ is the voltage which depends only on the supercapacitor capacitance aspect. Because of the variation of supercapacitor serial resistance R_1 with temperature, $V(t)$ increases with temperature decrease, the voltage $V_c(t)$ is directly proportional to the supercapacitor charge time Δt at constant current and for a given capacitance. For a given charge voltage, the serial resistance increases when temperature decreases, thus causing a decrease of the voltage $V(t) = R_1 I_{\text{ch}} + V_c(t)$ and hence the supercapacitor charge time decreases with temperature. The second effect is due to the decrease of the total supercapacitor capacitance with temperature. Indeed, the capacitance variations are due to variations of the activated carbon characteristics used in supercapacitor, of the electrical conductivity of the electrolyte, and of the double-layer effective thickness as a function of temperature [14]. These variations concerning supercapacitor serial resistance and capacitance have also been observed with 10, 50 and 100 A charge currents.

To quantify these variations, capacitance C_0 and C_v have been measured for different temperature values. Using the method described in Section 2 we have determined C_0 and C_v . Table 1 represents C_0 and C_v values as a function of temperature, the measurements have been done during constant current charge and discharge of a 3700 F.

Table 1
Parameter values for a 3700 F/2.5 V supercapacitor

	Temperature ($^\circ\text{C}$)			
	-35	-15	0	25
C_0 (F)	1979	2186	2255	2399
C_v (F/V)	672	638	611	553

Temperature variations of C_0 and C_v are less important than the serial resistance R_1 . For example, when the temperature varies from -35 to 25°C , the value of C_0 increases by about 21% and C_v decreases by about 17%. These variations are very low compared with R_1 variations.

In order to establish a relation between C_0 , and C_v and temperature, we represent in Fig. 6 their thermal variations. It can be seen that C_0 decreases when the temperature falls, whereas C_v slightly increases. However, since C_0 has the important value, the variation of the supercapacitor global capacitance with temperature depends principally on the thermal variation of C_0 .

We thus conclude that the supercapacitor global capacitance slightly decreases with temperature, as confirmed by Fig. 4. The consequence of the decrease of the supercapacitor capacitance with temperature is that energy storage in the supercapacitor is lower for low temperature values.

Using the experimental results, we have setup empirical laws for the variations of C_0 and C_v with temperature 3°C polynomial equations are sufficient to represent the experimental results.

The relations describe the evolution of C_0 and C_v versus temperature are, respectively

$$C_0(T) = a_3 T^3 + a_2 T^2 + a_1 T + a_0,$$

$$C_v(T) = b_3 T^3 + b_2 T^2 + b_1 T + b_0$$

a_i and b_i are determined from the fitted curves of C_0 and C_v . Table 2 gives a_i and b_i .

These relations are quite simple to implement in Saber and Spice software, simulation times remaining almost unmodified.

4.4. Simulation results

The electric equivalent circuit of supercapacitor, for which R_1 , C_0 and C_v take into account the temperature variations, has been implemented in the Saber and Spice software.

The simulation and experimental results for 2700 and 3700 F supercapacitor charge cycles are plotted in Fig. 7. Fig. 7a is the result for 3700 F supercapacitor. Simulation and experimental curves are obtained at 145 A of supercapacitor current charge, the temperature of the device is in order of -35°C . Fig. 7b represents the evolution of 2700 F

Table 2
Parameters of the polynomial fit of C_0 and C_v thermal variations

2700 F	Unit	Value
a_3	F ($^\circ\text{C}$) ⁻³	0.0032
a_2	F ($^\circ\text{C}$) ⁻²	-0.1251
a_1	F ($^\circ\text{C}$) ⁻¹	1.4572
a_0	F	1984.5
b_3	F ($^\circ\text{C}$) ⁻³	-0.0022
b_2	F ($^\circ\text{C}$) ⁻²	0.0491
b_1	F ($^\circ\text{C}$) ⁻¹	0.02470
b_0	F	543.70

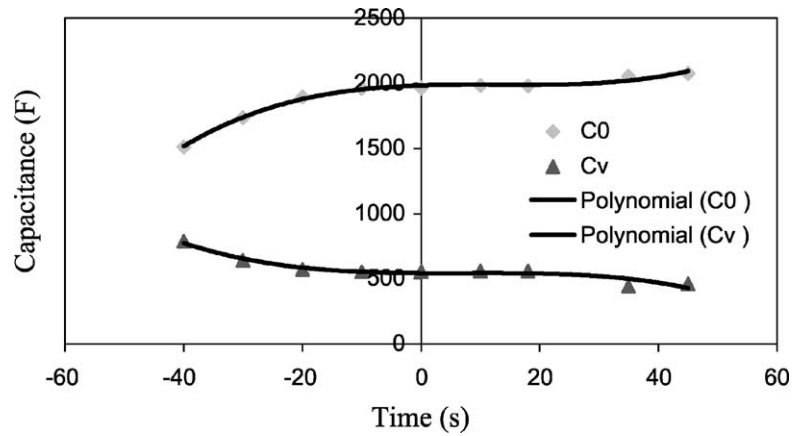


Fig. 6. Variation of C_0 and C_v vs. temperature for 2700F supercapacitor.

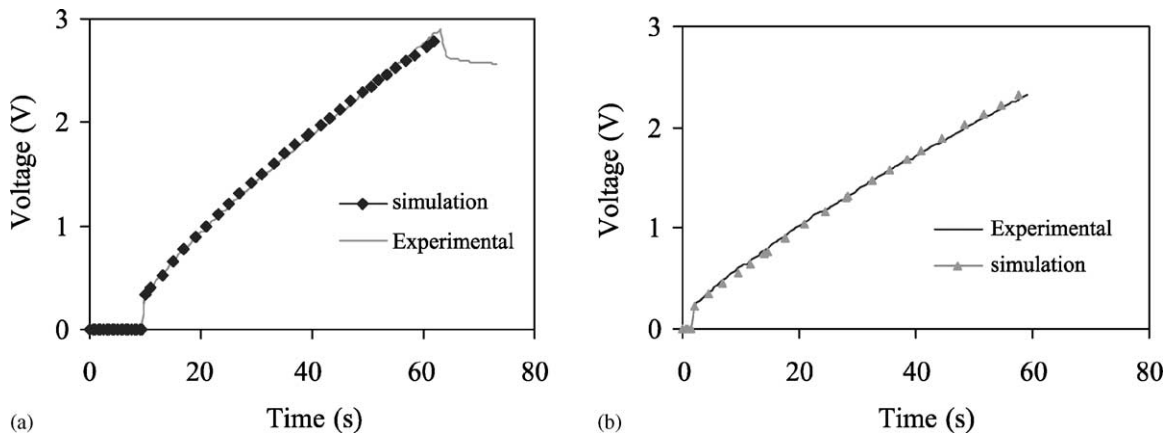


Fig. 7. Simulation and experimental curves for 3700F (a) and 2700F (b).

cycle charge at 95 A, the temperature is fixed in order of 35 °C.

It is clear that the two curves of simulation and experimental results for 3700F are similar. In the case of 2700 F, we can observe a very small difference between the experimental curve and the simulation one. This difference is due to the interpolation of the experimental curve by a polynomial law. However, in practice this error can be neglected. So we can conclude that the experimental results and the simulations ones are in good agreement. This result confirms the good behavior of the proposed electric equivalent circuit of supercapacitor.

The equivalent circuit of supercapacitor described in this paper, can thus describe the behavior of a supercapacitor as a function of temperature. It has been used to simulate supercapacitor packs in order to dimension the pack and to optimize their use in transportation applications.

4.5. Thermal management

Heat management problems with supercapacitor depend on the power level of operation and on the thermal environment of the device. We know that the optimum heat loss can

be achieved by maximizing surface-to-volume ratio. These two requirements are in opposition because supercapacitor store high levels of energy in a small volume and operate at high current level. The joule heating occurs from the current through the equivalent serial resistance must be evacuated. However, it is necessary to previous a heat sink. In Fig. 8 we

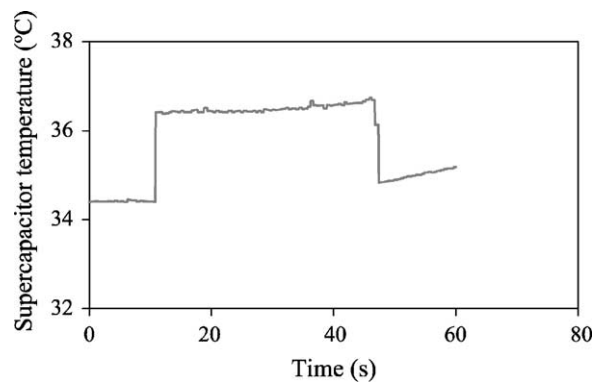


Fig. 8. Evolution of the supercapacitor surface temperature of 2700F supercapacitor, the current is in order of 140A and the temperature is in order of 35 °C.

have plotted the evolution of the temperature at the surface of 2700 F supercapacitor. This last is inside in the climatic room. The current of charge is in order of 140 A. This result shows that the supercapacitor surface temperature rises about 2 °C when the device operates. Moreover, to characterize the thermal behavior of supercapacitor for thermal management, it is necessary to determine the temperature profile inside the supercapacitor and in order to calculate the thermal resistance of the device. Results of this study will be published in the future.

For high voltage, it is necessary to use a pack of supercapacitor. The heat dissipation problem will be worse than for single cell units because of the high power losses. So, supercapacitor pack performances depend considerably of the thermal management of the pack especially in transportation applications. For example, in automotive applications, a power source 42 V is obtained by using 18 cells of 2700 F supercapacitor. If the supercapacitor current is in order of 100 A, the power dissipated is approximately 180 W at 20 °C. However, it is necessary to evacuate this heat power in the aim to have a good efficiency of supercapacitor.

5. Conclusion

We showed that supercapacitor ESR increases when temperature decreases. The capacitance decreases with temperature. Consequently, of these results, the power dissipated increases and the energy stored in the supercapacitor decreases with temperature. So, the supercapacitor efficiency decreases.

An equivalent electric circuit of 2700 and 3700 F supercapacitors is realized by taking into account the thermal variations of the device and its environment. The electric equivalent circuit proposed will be easy to use in several analog simulators. It is well adapted to some studies concerning behavior of supercapacitors when used in automotive applications thanks to sensitivity analysis induced by thermal parameters. The simulated results obtained are in good agreement with the experimental ones.

Finally, heat management problems with supercapacitor are discussed, and the supercapacitor performances in transportation applications depend considerably of the thermal management.

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