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Lithium-Ion Capacitor - Advanced Technology for Rechargeable Energy Storage Systems

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Abstract

This paper presents the electrical and thermal behaviour of an advanced lithium-ion capacitor (LIC) based rechargeable energy storage systems. In the proposed study, an extended statistical analysis has been performed to evaluate the main electrical parameters such as resistance, power, capacitance, rate capabilities, variation between cells and thermal parameters. Based on the performed analysis, an electrical model has been developed for dimensioning and evaluation of various applications based on lithium-ion capacitor technology.

Keywords: Lithium-Ion Capacitor, EDLC, Lithium-Ion Battery, HEV, BEV

1 Introduction

Since the beginning of the automobile era, the internal combustion engine (ICE) has been used for vehicular propulsions. In addition, motor vehicles powered by the ICE are significant contributors to air pollutants and greenhouse gases linked to the global climate change [1,2]. As the global economy begins to strain under the pressure of rising petroleum prices and environmental concerns, research has spurred the development of various types of the clean energy transportation systems such as Hybrid Electric Vehicles (HEVs), Battery Electric Vehicles (BEVs) and Plug-in Hybrid Electric Vehicles (PHEVs) [3,4]. However, the establishment of the energy storage technology, which can support the output power during acceleration, the efficient use of the regenerative energy and considerable life cycle, are the critical aspects. Furthermore, there is no current battery technology can meet these often concurrent objectives [4-12].
A possible solution for enhancing the present battery performances is the hybridization of batteries with electrical double-layer capacitors (EDLCs) [13-14]. However, such hybrid architecture needs expensive and high efficient DC-DC converters. This makes this combination less attractive in mobile applications where cost, weight and volume can be considered as main barriers [14].
In order to overcome these obstacles, in the last decade, several types of advanced rechargeable energy storage systems have been developed by many companies and research centers (called lithium-ion capacitors or hybrid capacitors) [15-20]. In [21-24], the general performances of these technologies have been investigated. In this study, they observed that an energy density of 14 Wh/kg and over 10000 W/kg could be achieved. In [25-28], the suitability of the proposed technology has been analyzed. From these works, it can be seen that lithium-ion capacitors can act as a main power or energy source without any reduction of the system capabilities.
Furthermore, the LIC seems in certain sense more attractive than EDLCs due to their higher operating

conditions (such as voltage 3.8V max. compared to 2.7V for EDLCs). This means that the needed cells that are connected in series will be lower, and the complexity of the entire system will be reduced.

2 Working mechanism lithium-ion capacitor

The Lithium-Ion Capacitor is a rechargeable energy storage system, which belongs to the class of hybrid capacitors or asymmetric capacitors. It can be classified between lithium-ion batteries and electrical-double layer capacitor (EDLC) The positive electrode uses porous activated carbon as in conventional EDLCs. The electrode has been prepared by carbonized of precursors. The specific capacitance of the electrode is about 100F/g, which is assumed based on 1000 m^2/g for the surface area of the electrode and 0.1 F/m^2 for the double-layer [14]. The negative electrode also uses carbon material with significant Li-ion pre-doped with lithium-ion on its negative electrode (see Figure 1) [1,6-10]. The used electrolyte is an organic based carbonate mixture. The key technology is the pre-doping of the Li to the anode carbon for enhancing the energy density. The Li foil is set close to the assembled electrodes, which are supported by porous current collectors and is connected with porous current collector of the anode. After impregnation of electrolyte, Li pre-doping proceeds by dissolving Li into the electrolyte and moving into anode to low the potential of anode carbon. The anode potential is lowered by charging process of the LIC. The potential of the negative electrode can be kept low during discharge of the cell since the capacity of anode is significantly larger than that of cathode [11]. Figure 1 shows the elementary structure of lithium-ion, EDLC and Li-ion capacitor structure.

It can be seen that the negative LIC electrode is formed by Li doped carbon. The LIC equivalent capacitance is formed by the positive electrode capacitance C^+ in series with the negative one C^-. The equivalent capacitor can be expressed as following:

$$\frac{1}{C_{cell}} = \frac{1}{C-} + \frac{1}{C+} \qquad (1)$$

Since C^- is much higher than C^+, the capacitance of the LIC cell C_{cell} is nearly equal to the capacitance of the activated carbon-electrolyte C^+. Here, it should be noted that occur of thermal runaway is limited since the cathode material is composed of activated carbon as in conventional EDLCs.

In this paper, the prismatic lithium-ion capacitor cells 3300F fabricated from JM Energy have been investigated. The main characteristics of this LIC type are summarized below:

- Nominal capacitance: 3300F,
- Weight: 350 g,
- Maximum voltage: 3.8 V,
- Minimum voltage: 2.2V,

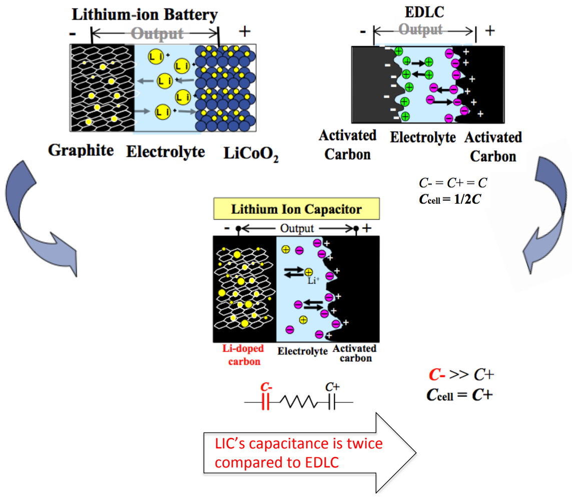

Figure 1: Comparison working mechanisms of EDLCs, lithium-ion batteries and LIC

3 Applications & Requirements

In the last decade, EDLCs have been implemented in many applications where the peak power is needed. However, the energy content of the EDLCs is not enough whereby several parallel stacks are required. From this context, the use of lithium-ion capacitors could be an interesting solution in vehicular applications where still high peak powers with higher energy content than EDLCs are desired. Furthermore, the selection of the investigated prismatic LIC has the advantages in terms of compact integration in a pack. In addition, the prismatic shape offers significant space advantage, which can be considered of high importance in mobile applications such as buses, trams and metros.

Application	Voltage [V]	Power [W]	Cycle life	Duration
Bus	700-800	150k	1000.000 cycles	10 s
Metros	800-900	1-2M	200.000 – 400.000 cycles (1 year)	10 – 20 s
Trams	700	300k-400k	?	10 – 20 s
Load leveling	400	200k – 1G	2500 (10 years)	50 – 300 min
Back-up power	400	1k – 1M	100 (10 years)	15 min
Cranes	800	350k	1000.000 cycles	10 s

Table 1: Specifications of some applications

4 Test methodology

In this study, a methodology as presented in Figure 2 has been used for investigating the performances of the proposed LIC cells. As one can observe the methodology consists of electrical, thermal and electrochemical impedance spectroscopy (EIS) tests.

The proposed methodology will assist us to have the required insight of the LIC performances in terms of energy, capacity, power, thermal and EIS performances at different working conditions such as current, ambient temperature and state of charge (SoC).

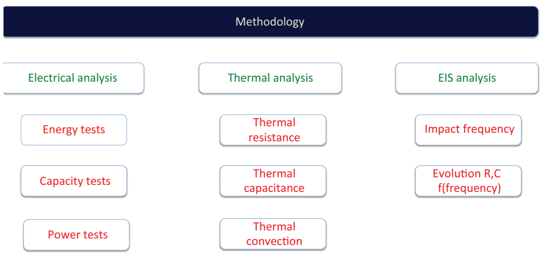

Figure 2: Test methodology

5 Results

5.1. Energy test

In Figure 3, the energy density evolution of the LIC is demonstrated. The energy test has been conducted at 10A charging until maximum voltage (3.8V) following by constant voltage until the current was reduced to 0.01A. After a rest period of 15 minutes the cells have been discharged at different current rates till 2.2V, which is the minimum voltage. Prior starting the tests, the cells have been placed in a climate chamber at the desired temperature for a period of 3 hours.

As one can observe the energy density of the LIC at 10A discharge is between 12.1 Wh/kg and 11.4 Wh/kg at ambient temperatures between 60°C and -10°C, respectively. Then, the energy density decreases slightly as a function of the applied current. At 200A, the energy density is between 10.2 Wh/kg and 6.6 Wh/kg.

In [24], the authors documented that the increase of the energy density of the EDLCs is less dependent on the above-mentioned parameters. The reason is attributed to the pseudo-capacitance reactions that occur in the LIC. In [29], the authors reported that the decrease of the energy in the LIC at low temperatures is due to the increase of the electrolyte resistance. Here one can conclude that the role of the thermal management is vital for LIC for keeping the cells or the system in the appropriate operating window, where high performances can be assured.

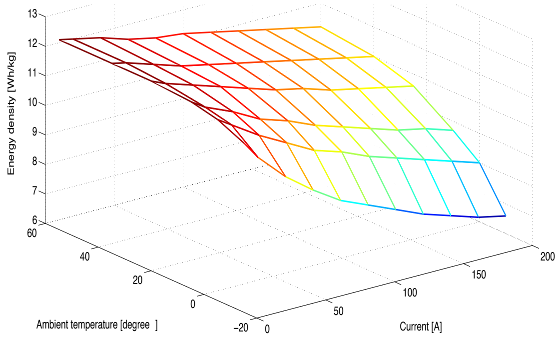

Figure 3: Energy density evolution versus current rate and ambient temperature

5.2. Capacity test

In load levelling and back-up power applications, the LIC should to be able to supply the required capacity or power during a specific duration. According to the specifications as can be found in Table 1, the operation duration could be between 15 and 300 minutes.

Therefore, in Figure 4, the capacity evolution has been obtained from the previous test at different current rates and ambient temperatures. As we can see, the capacity decreases more pronounced compared to EDLC as function of current rates and ambient temperature. The obtained evolution

is similar to the high power lithium-ion batteries such as lithium titanate oxide based [8].
This evolution predicts that the Peukert number for the LIC is higher than 1 as can be seen in Figure 5. In Figure 5 one can see that the Peukert number changes between 1.02 and 1.11 in the temperature range 60°C and -10°C.
Peukert phenomenon is an empirical relationship, which describes the rate capabilities of the cell at different current rates. The general equation of the Peukert relationship is presented by equation (2):

$$C_{dis} = T.I^k \tag{2}$$

where C_p is the discharge capacity in Ah, while T is the time in second and I represents the discharge current, respectively. k is the Peukert constant, and equals to one for an ideal rechargeable energy storage systems where the capacity is almost equal independent of the employed current.

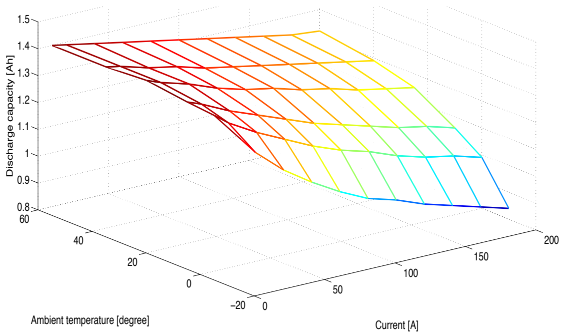

Figure 4: Evolution of the discharge capacity as function of current rate at different ambient temperatures

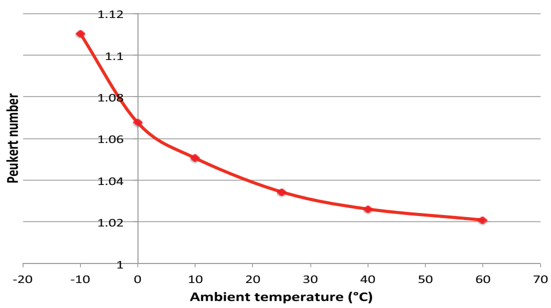

Figure 5: Evolution of Peukert number versus ambient temperature

5.3. Power capabilities

Besides energy, power requirements can be considered as a key criterion in the application of LIC. In most applications, the LIC should to have high power capabilities during discharge and charge as well.
Therefore, in this paper, an extended analysis has been performed to evaluate the power performances of the proposed LIC at different SoC (from 0% to 100% with steps of 5%), at current rates (20A to 200A with steps of 20A) and at the same working temperatures as in sections 5.1 and 5.2.
The power capabilities have been investigated based on 1 second pulses during charging and discharging.
In Figure 6, the experimental results of the power density versus state of charge during one-second pulse at different working temperatures are illustrated. The power density has been calculated based on the equation (3).

$$P_{density} = \frac{U_{rated}^2}{4 \cdot m \cdot R} \tag{3}$$

Where,
$P_{density}$: power density [W/kg],
U_{rated}: the voltage at specific SoC [V],
R: internal resistance of LIC during the pulse [Ω],
m: mass of the LIC cell [kg],

As we can observe the power density of the LIC is strongly depending on the working temperature. As one can notice that the power density (at 60°C, 100% SoC) is 9000 W/kg against 3170 W/kg (at -10°C). The decrease of the power at lower temperatures is attributed to the increase of the electrolyte resistance [24]. From Figure 6, we can conclude that LIC cells are able to supply peak powers at lower SoC levels as well. However, the peak powers at higher SoC levels are significantly higher.
Here it should be noted that the applied current was only limited to 200A. However, from Figure 7, we can observe that the voltage drop at high SoC levels is not big, which means that higher current rates can be applied. Since the resistance of the LIC decreases as a function of current, the power density will increase significantly at higher current rates.
Here it should be noted that the LIC cells need a thermal management to keep the cells in the appropriate operating conditions where high performances can be guaranteed.

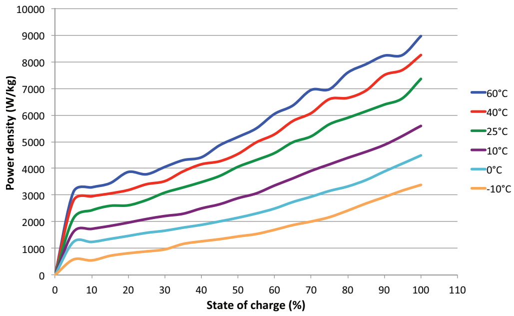

Figure 6: Discharge power density versus state of charge

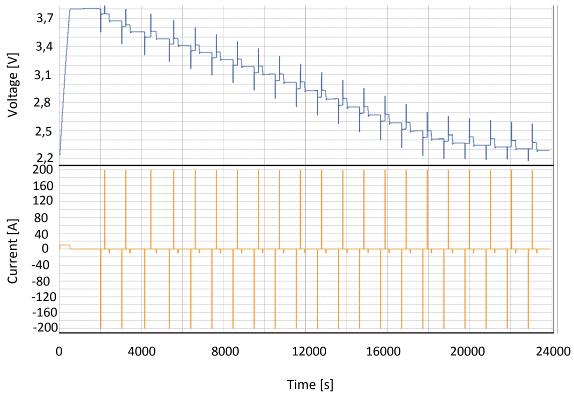

Figure 7: Representation of the voltage and current versus time during HPPC tests at 25°C and 200A

5.4. Impact frequency

In the field of energy storage testing, the electrochemical impedance spectroscopy can be considered as of high importance. Based on the evolution of the impedance as a function of frequency, some additional information can be obtained, which is only possible by using the battery tester.

In Figure 8, the evolution of the EIS measurements at different voltage levels is illustrated. As we can observe, in Figure 8, at 3.8V the bulk resistance is about 0.74mΩ.

In [23], the authors indicated that the series resistance of LICs consists of the bulk resistance, the charge transfer resistance and the diffusion resistance. The bulk resistance R_b represents the intersection with the real axis. The charge transfer resistance R_e is the half circle, while the diffusion resistance starts when the resistance R_e ends. However, from Figure 8, the charge transfer resistance is difficult to observe.

It is clear that there is a shift of the bulk resistance over the real axis when the cell voltage changes. Generally, one can conclude that the resistance R_e increases when the voltage decreases.

Another phenomenon that can be noticed at low frequencies is the shift of the spectra as function of the rated voltage.

This indicates that at low frequencies the LIC behaves as a capacitive cell. This observation also can be approved by Figure 8, where the phase at 10 mHz is about -80°. Here it should be noted that the differences in phase at different voltage levels are small. However, the variations at 10 kHz are high.

Furthermore, in Figure 10, one can notice that the appropriate frequency range is between 0.1 Hz and 1 kHz. This is due to the smaller impedance.

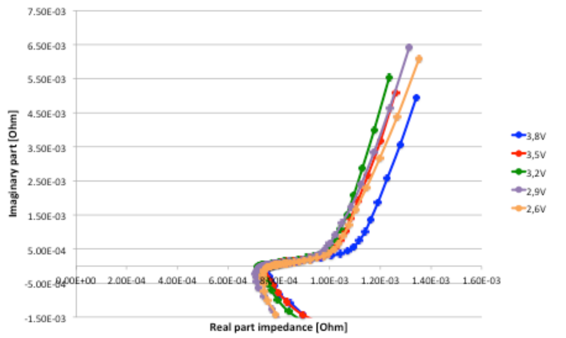

Figure 8: Nyguist plot at different voltage levels

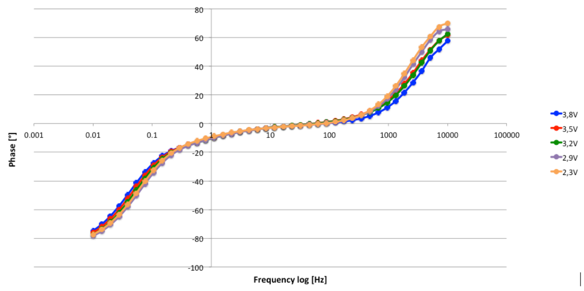

Figure 9: Phase evolution versus frequency at different voltage levels

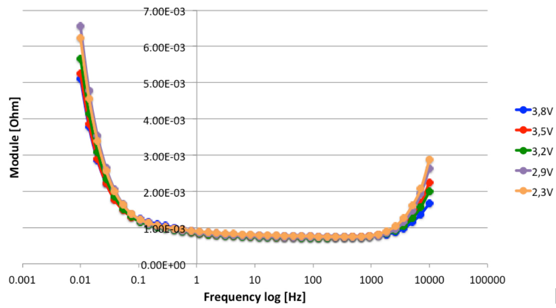

Figure 10: Module evolution versus frequency at different voltage levels

Since the capacitance of the LIC is depending on the voltage, there is a need to have a clear view of the capacitance performances as a function of voltage level. In Figure 11, the capacitance of the

LIC at 25°C and different voltage levels is demonstrated. As one can notice, the capacitance is clearly varying as a function of voltage in the frequency range 10 mHz and 1Hz. Here it should be underlined that the capacitance at 2.6V is higher than at 2.9V. The reason for this result can be explained by the higher increase of the imaginary part of the impedance at 2.9V against at 2.6V as can be seen in Figure 8.
The capacitance of the LIC has been calculated based on the equation (4):

$$C = \frac{-1}{2 * \pi * f * Z(f)} \tag{4}$$

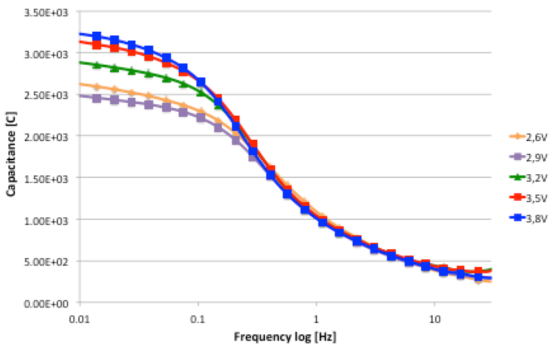

Figure 11: Capacitance versus frequency at different voltage level

5.5. Thermal behaviour

Prediction of thermal behaviour of energy storage systems can be considered nowadays of high importance in the scientific community. The temperature of the cell in real applications should be monitored in order to avoid critical situation like thermal runaway. Then, the performances of cells are strongly temperature dependent, which indicates that the need of a thermal management is high.
Therefore, accurate thermal models could be interesting tool to examine the thermal behaviour of LIC cells in advance.
In this paper, a test profile as illustrated in Figure 12 has been used. The profile exists of charge and discharge pulses at 100A. The pulse durations are 3s with 1s rest time between charge and discharge. The proposed test has been carried out at 25°C. From this test, one can observe that the temperature increases until the temperature is stabilized.
Based on such evolution, the thermal parameters such as thermal resistance R_{th}, thermal convection R_{con}, thermal capacitance C_{th} and thermal time constant τ_{th} can be determined.

The thermal convection, which describes the heat exchange between the cell surface and the environment, has been calculated based on the equation (5):

$$R_{con} = \frac{T_a - T_{surface}}{R \cdot I^2} \tag{5}$$

where
R_{con}: the thermal convection [°C/W],
T_a: the ambient temperature [°C],
T_s: the surface temperature of the cell [°C],
R: the internal resistance of the cell [Ω],
I: the current through the cell [A],

The calculated thermal convection is 1.42 °C/W.

Since the evolution of the temperature is a first order, the thermal time constant is 66% of the value when the temperature stabilizes. In this case, the time constant is 1300 s.
In literature, it is generally known that the thermal resistance of such technology is about 3-3.2°C/W [30].
Based on those parameters, the thermal capacitance can be determined as follows:

$$\tau_{th} = C_{th} \cdot (R_{th} + R_{con}) \tag{6}$$

According to the calculated parameters, the thermal capacitance is 294 J/°C.

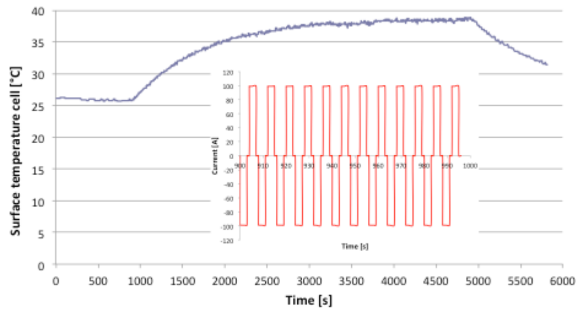

Figure 12: Temperature evolution during cycling the LIC cell at 25°C & 100A

5.6. Voltage characteristics

Since lithium-ion capacitors are emerging rechargeable energy storage technology, there is up to now no clear answer if they belong the EDLCs or lithium-ion technology. In order to answer on this question, the voltage response during one second pulse at 100A discharge and at

25°C as demonstrated in Figure 13 has been investigated in depth.
In the case of EDLCs, the voltage response during a pulse reveals an immediate voltage drop, which stands for the equivalent series resistance (ESR) as indicated in the standard IEC 62576 [31]. Then, the voltage decreases linearly. However, Figure 13 reveals that the voltage shows an immediate voltage drop, which indicates the ohmic resistance of the LIC. Then, the voltage decreases exponentially till the end of the pulse, which represents the polarization behaviour. As indicated in [8], this evolution is comparable to lithium-ion batteries.
From this point of view, one can conclude that LIC belong the lithium-ion group, but with very high power capabilities. Thus, the standard IEC 62660-1 also can be applied on LICs [32].

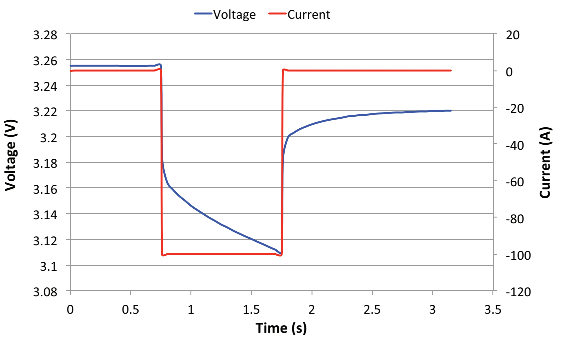

Figure 13: Voltage evolution of the LIC at room temperature during a pulse of 1 s at 25°C

6 Lithium-ion capacitor model

As demonstrated in Table 1, LICs can be implemented in many applications. In order to make the use of this energy storage system possible, there is a need to have an accurate electrical model, which is able to predict the LIC performances during real operation of the system.
Therefore, in this paper, the modified FreedomCar first order model has been used for this purpose as shown in Figure 14. In order to increase the model accuracy, the hysteresis has been taken into account by separating the charge and discharge ohmic resistances and polarization circuits.
The LIC model parameters have been estimated based on an advanced minimization technique at VUB. Therefore, the HPPC sequence has been conducted at different SoC (from 100% to 0% with steps of 5%), ambient temperatures (60°C, 40°C, 25°C, 10°C, 0°C and -10°C) and current rates (20A, 40A, 60A, 80A, 100A, 120A, 140A, 160A, 180 and 200A).

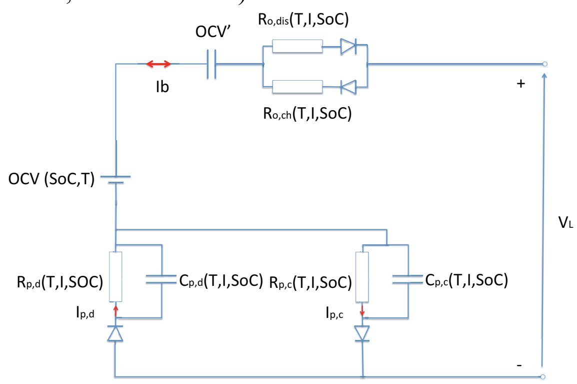

Figure 14: New electrical model for lithium-ion capacitor

In order to evaluate the accuracy of the new electrical model, two different load profiles have been selected. The first test is the discharge tests at 10A and 25°C from 3.8V till 2.2V as shown in Figure 15. One can observe that the model is in good agreement with the experimental results. The maximum error percentage is around 0.25%.
The second test is the dynamic discharge performance test as documented in the standard IEC 61982-2 [33]. In Figure 16, one can notice that the high performances of the model can be achieved when a dynamic load profile is applied. The error percentage between the model and the experimental result is about 1.5%. The error percentage has been calculated based on equation (7).

$$Error = \frac{Voltage_{model} - Voltage_{experimental}}{Voltage_{model}} \cdot 100\% \qquad (7)$$

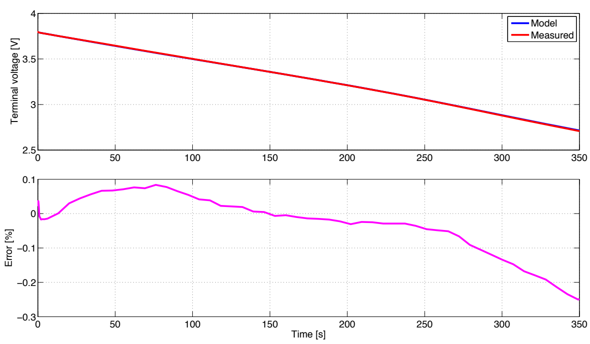

Figure 15: Comparison of the experimental and simulation based on constant 10A discharging at 25°C

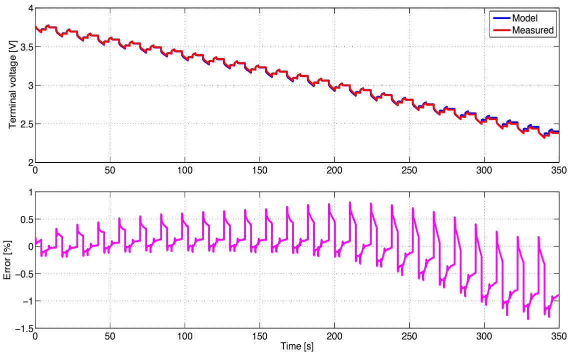

Figure 16: Comparison of the experimental and simulation based on DDP test at 25°C

7 Summary and conclusions

In this paper, an extended analysis has been performed of the performances of lithium-ion capacitors at different working conditions. Then, the characteristics of this technology have been investigated and we observed that lithium-ion capacitors could be categorized into high power lithium-ion batteries.

Furthermore, the experimental results showed that LIC could be an interesting candidate to replace EDLCs. Particularly, the combination of high power and high energy density can provide a better performance compared to the EDLCs in many applications. However, this technology offers advantages in terms of integration possibilities.

Finally, this paper proposes an enhanced electrical model, which can predict LIC behaviour accurately.

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