Electrochemical double layer capacitor and lithium-ion capacitor based on carbon black

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ABSTRACT

In this paper we report the physical investigation and the electrochemical performance of the carbon black SC3 from Cabot Corporation. The SC3 carbon black was investigated in terms of BET surface area, pore size distribution, resistivity and morphology. Composite electrodes containing SC3 as active material were prepared and used for the realization of electrochemical double layer capacitor (EDLC) and lithium-ion capacitor (LIC). In EDLC, at $5\,\mathrm{mA\,cm^{-2}}$ charge–discharge currents, the carbon black displays a specific capacity of $40\,\mathrm{mAh\,g^{-1}}$ and a specific capacitance of $115\,\mathrm{F\,g^{-1}}$. It also displays a very good cycling stability for over 50,000 cycles and excellent performance retention at currents up to $50\,\mathrm{mA\,cm^{-2}}$. The performance retention at high currents outstandingly differentiates this carbon black from a few commercially available EDLC-grade activated carbons. Because of the high specific capacity of SC3, the carbon black electrodes were also used in combination with $LiFePO_4$ electrodes in LIC. The results of this study indicate that SC3 carbon black is an interesting carbonaceous candidate for the realization of LIC.

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1. Introduction

Carbon based electrochemical double layer capacitors (EDLCs), also known as supercapacitors or ultracapacitors, are advancing as one of the most promising energy storage technology. In EDLCs the charge is electrostatically stored at the carbon–electrolyte interface and, because of this storage mechanism, EDLCs can be charged–discharged in seconds. EDLCs normally display high specific power and they also have an extremely high cycle life (>500,000). Because of these characteristics, EDLCs are conveniently used in a large number of applications where rapid charge–discharge capability and reliability are required [1–3].

At the moment, considerable research is focused on improving of the specific energy of EDLCs, which is defined by the expression $E = (1/2)CV^2$, where C and V are the capacitance and operating voltage of EDLC. Introduction of novel or improved carbonaceous materials with high capacitance, e.g. carbon nanotubes [4–8], and realization of systems with high operating voltage, e.g. using ionic liquids as electrolytes [9–11], are the two distinct approaches for energy improvement of EDLCs. Recently, another approach of combining a typical lithium-ion battery electrode with an EDLC carbon-based electrode has been proposed as high energy stor-age solution, also known as lithium-ion capacitor (LIC) [12–15]. Because of the combination of different types of electrodes, LIC can have energy and power densities in the range between those of lithium-ion batteries and EDLCs, and, at the same time, also display an improved cycling stability compared with batteries.

Carbon blacks are currently used in EDLCs, but primarily as conductive additives to activated carbon electrodes at 5–10 wt.% loading. In this application, carbon blacks usually have the dry powder conductivity in the range of $10\,\mathrm{S\,cm^{-1}}$. Carbon blacks have a fractal-like structure of many nano-sized primary particles of around 10 nm in diameter that are fused together within aggregates of typically 100 nm in size. Produced by partial combustion or thermal decomposition of hydrocarbons (usually gases, oils, or distillates) in the gas phase [16,17], the properties of carbon black can be controlled by the manufacturing process. For example, the surface area of carbon black may be engineered from around $10\,\mathrm{m^2\,g^{-1}}$ to an excess of $1500\,\mathrm{m^2\,g^{-1}}$. Interestingly, the surface of carbon black is generally considered to be more accessible than that of other forms of high surface area carbons [18]. For this reason, carbon blacks have been proposed and used as the sole active material in EDLCs, thus replacing the activated carbons [18–23]. However, due to high amount of binder necessary for the realization of mechanically stable electrodes and poor packing density of commercially existing carbon blacks, the carbon black performance did not favourably compete with systems based on activated carbons.

In this paper, we investigate the performance of the carbon black SC3 from Cabot Corporation as active material in EDLCs. In the first

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part of the paper, the physical characterization (BET, particle size, resistivity) of SC3 is reported. In the second part, the performance of carbon black is investigated in terms of specific capacitance, capacity and cycle stability. Finally, taking into account the observed high specific capacity of SC3, we report on the performance of this material in combination with lithium and LiFePO$_4$ counter electrodes in LIC.

The results of our study indicate that the carbon black SC3 is an attractive active material for the realization of EDLCs and, moreover, the development of lithium-ion capacitors.

2. Experimental

Carbon black SC3 (Cabot Corporation), activated carbon DLC Super 30 (Norit), sodium carboxymethyl-cellulose (CMC, Walocel CRT 2000 PPA 12 from Dow Wolff Cellulosics) and carbon-coated LiFePO$_4$ (Südchemie) were used as received in electrode preparation. Carbon black Super-P (TIMCAL) was used as conductive agent.

For the preparation of SC3 electrodes, CMC was dissolved in water to obtain a 1.5 wt.% solution and equilibrated for 2 h at room temperature with a magnetic stirrer. The active material (SC3) and conductive agent (Super-P) were mixed together for about 2 h. Then, the material was added to the solution containing CMC and dispersed with an high energy stirrer (Ultra Turrax, IKA) for 1 h. The so-obtained slurry was casted immediately on aluminium foil (30 μm, purity > 99.9%, etched by immersion in 5 wt.% KOH at 60 °C for 60 s) by using a laboratory scale doctor blade coater, whose blade was set at 100 μm. The coated foil was dried in an ambient atmosphere oven at 80 °C over night. After drying, the electrodes with a diameter of 12 mm were stamped from the coated sheets. Finally, the prepared electrodes were dried in a glass oven under vacuum at 170 °C for 24 h. The composition of the dried electrode was 70 wt.% SC3, 20 wt.% Super-P and 10 wt.% CMC. The average mass loading of the electrodes was about 0.7 mg cm^{-2}.

We also compared the performance of SC3 with that of commercially available activated carbon (AC). We used an EDLC grade of AC from Norit, DLC Super 30. For preparation of AC electrodes, a procedure identical to that described above was used. The composition of the dried electrode was 80 wt.% AC, 15 wt.% Super-P and 5 wt.% CMC. The average mass loading of the electrodes was about 1.5 mg cm^{-2}.

All electrochemical tests were carried out with 3-electrode Swagelok®-type cells. The cells were assembled in an Argon-filled glove box with oxygen and water contents lower than 1 ppm. In the first series of experiments, a SC3 electrode was used with a carbon counter electrode, whose mass loading was about 4 mg cm^{-2}. In the second series of experiments, two identical SC3 and DLC Super 30 electrodes were assembled in an EDLC. Both experiments were carried out using an Ag wire as quasi reference electrode and 1 M Et$_4$NBF$_4$ in propylene carbonate (PC) as an electrolyte solution. In the third series of experiments, SC3 and DLC Super 30 electrodes were used against metallic lithium electrode (metallic lithium from Chemetall). In the fourth series of experiments, SC3 electrode was used against LiFePO$_4$ electrode in a LIC. The preparation of LiFePO$_4$ electrode is described elsewhere [24]. In the last two series of experiments, lithium metal was used as reference electrode and 1 M LiPF$_6$ in PC was used as electrolyte solution. In all experiments a Whatman GF/D glass microfiber filter of 675 μm in thickness and 12 mm in diameter was used as a separator. It was drenched with 80 μL of electrolyte solution.

All electrochemical tests were performed at room temperature using a VMP multichannel potentiostatic–galvanostatic system (Biologic Science Instrument, France). Cyclic voltammetry was carried out at scan rates ranging from 5 to 200 mV s^{-1}. Galvanostatic cycling measurements were carried out using currents ranging

from 5 to 50 mA cm^{-2}. The capacitance (C), capacity (Q), equivalent series resistance (ESR) and efficiency (η) were obtained by applying Eqs. (1)–(4) to the voltage profiles, which were measured at certain charge–discharge currents. In the equations, s is the slope of discharge curve, i is the current, m$_{act}$ is the active mass loading of one electrode, t$_d$ is the discharge time, t$_c$ is the charge time, ΔV is the ohmic drop and A is the electrode surface area.

$$C\,[\mathrm{F\,g^{-1}}] = \frac{i\,[\mathrm{A}]}{s\,[\mathrm{V\,s^{-1}}]m_{act}\,[\mathrm{g}]} \tag{1}$$

$$Q\,[\mathrm{mAh\,g^{-1}}] = t_d\,[\mathrm{s}]\frac{i\,[\mathrm{A}]}{3.6m_{act}\,[\mathrm{g}]} \tag{2}$$

$$\eta\,[\%] = \frac{t_d\,[\mathrm{s}]}{t_c\,[\mathrm{s}]} \times 100 \tag{3}$$

$$\mathrm{ESR}\,[\Omega\,\mathrm{cm^2}] = \frac{\Delta V\,[\mathrm{V}]}{2i\,[\mathrm{A}]} \times A\,[\mathrm{cm^2}] \tag{4}$$

The nitrogen BET surface area was measured according to ASTM Standard D6556. The oil absorption number (OAN) and crushed oil absorption number (COAN) are also known as DBP (dibutyl (n-dibutyl) phthalate) and crushed DBP (CDBP) adsorption numbers, respectively. The OAN and COAN are the measures of carbon black structure, the latter is determined after the controlled compression of carbon black, having the units of mL of DBP oil adsorbed per 100 g of carbon black. The OAN and COAN are measured based upon ASTM Standard D3493-06. The dry powder resistivity is measured following the procedures described by Espinola et al. [25]. The carbon black primary particle size is measured by statistical transmission electron microscopy (TEM) analysis.
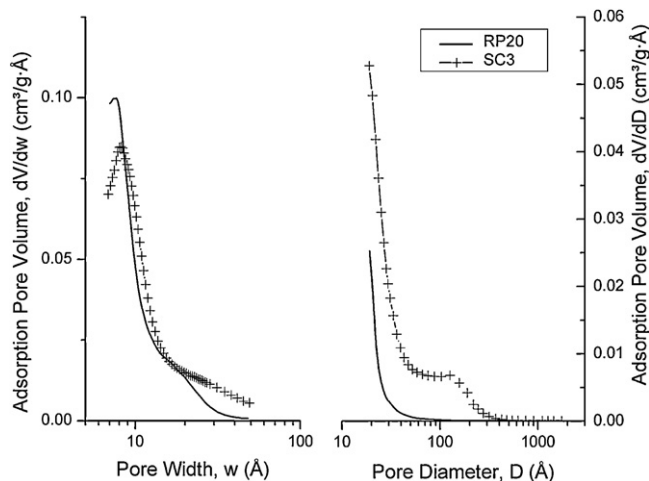
3. Results and discussion

3.1. Physical characterization and electrode preparation

The physical properties of the carbon black SC3 are given in Table 1 and compared with a commercially available carbon black of high surface area, Black Pearl 2000 (BP2000) from Cabot Corporation. As indicated in the table, SC3 displays higher surface area and higher resistivity with respect to BP2000. The oil absorption number, also known as DBP number, is lower for SC3 than for BP2000, indicating a higher packing density for SC3. It is interesting to note that the higher surface area and packing density of SC3 powder in comparison to those of BP2000 can also be thought in terms of improvement in volumetric and gravimetric capacitance of SC3. Since the pore size distribution can strongly affect the EDLC performance, we compared the pore size distribution of SC3 with commercially available supercapacitor grade activated carbon RP 20 from Kuraray Chemicals. The surface area of RP 20 is in the 1800 m^2 g^{-1} range. The comparison is shown in Fig. 1 and indicates a good match of SC3 to commercial materials. The observed pores of carbon black in the 10 nm range are due to voids between the primary particles. Moreover, due to smaller carbon black particle size in comparison to that of activated carbons, the length of pores within carbon black particles is shorter than that within activated carbons.

Fig. 2a shows an SEM image of the pristine SC3 powder. As shown in the figure, the carbon black displays a particle size ranging from 2 to 10 μm, with the majority of particles in the 2–4 μm range. The particles observed in this image consist of agglomerated carbon black particles, which, depending on the slurry mixing conditions, may or may not be altered for electrode preparation. Fig. 2b shows an SEM image of a SC3 electrode used in this work. It is important to note that the preparation of electrodes containing conventional activated carbons is normally less elaborate compared to that containing carbon black. This may be due to the intrinsic characteristics

Table 1
Physical properties of SC3 experimental carbon black in comparison with BP2000 commercially available carbon black.

	N2 BET surface area ($m^2 g^{-1}$)	Oil absorption number (mL/100 g)	Crushed oil absorption number (mL/100 g)	Powder resistivity at 20 MPa ($m\Omega$ cm)	Primary particle size (nm)
SC3	1880	255	215	330	8.4
BP2000	1500	330	280	208	12

Fig. 1. Comparison of pore size distribution of SC3 carbon black with RP 20 from Kuraray Chemicals.

Fig. 3. Specific capacitance values of SC3 carbon black electrode in PC-Et$_4$NBF$_4$ (1 M) obtained from CV at 20 mV s^{-1} by dividing the specific current by the scan rate. *Inset*: specific capacitance of SC3 carbon black electrode at different scan rates.

of carbon black, e.g. particle size, morphology, functional groups, etc. Nevertheless, by using 10 wt.% of CMC binder, it was possible to prepare mechanically stable electrodes containing the carbon black SC3. Moreover, as shown in Fig. 2b, the carbon black SC3 electrodes displayed an acceptable material distribution over the current collector and a quite homogeneous surface. Further optimization of electrode quality and packing density may be considered, but for the purposes of this work was not attempted.

3.2. EDLC based on SC3 carbon black

Fig. 3 shows the result of the first series of experiments, where SC3 electrode was used with a carbon counter electrode. Here, a cyclic voltammogram (CV) was carried out at various scan rates in an overall range of potential of 2.5 V (from −0.95 V to 1.55 V vs. Ag). An example of CV at 20 mV s^{-1} is shown. From the CV, the SC3 electrode displays a typical butterfly shape with no significant Faradaic peaks, which is a characteristic of capacitive material. The resultant capacitance of SC3 material is around 115 F g^{-1}, while the coulombic efficiency of the charge–discharge process is 100%. At higher scan rates, the specific capacitance of SC3 only slightly

decreases (see inset of Fig. 3), but the coulombic efficiency of the charge–discharge process stays at 100%.

Following the above results, the second series of experiments was carried out, where two SC3 electrodes were investigated in an EDLC configuration in the same voltage range which was used for CV tests (corresponding to an operative maximum voltage of 2.5 V). The EDLC was charged–discharged at several current densities, ranging from 5 to 50 mA cm^{-2}, for 50,000 cycles. As shown in Fig. 4, the performance of SC3 displays 100% efficiency of the charge–discharge process during all cycles and is independent on the current density. The ESR of the electrode was constantly lower than 2 Ω cm^2 after the initial fluctuation. At 5 mA cm^{-2}, the specific capacity and capacitance of the electrode are around 40 mAh g^{-1} and 115 F g^{-1}, respectively. For comparison, the performance obtained with an EDLC containing activated carbon (AC) was also investigated. The used activated carbon was DLC Super 30 (Norit), which has a surface area of 1400 m^2 g^{-1} and an average particle size of 6.3 µm. Using this AC-based EDLC, charge–discharge tests were carried out in conditions identical to those applied for the SC3-based EDLC. At 5 mA cm^{-2}, the AC electrode displays a
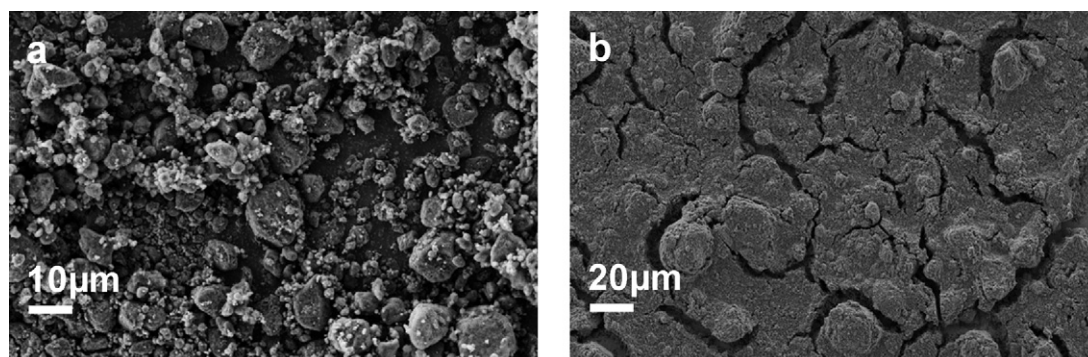
Fig. 2. (a) SEM images of pristine SC3 carbon black powder. (b) SEM images of a SC3-based electrode produced by coating from aqueous slurry (no calendaring or further electrode optimization was done).
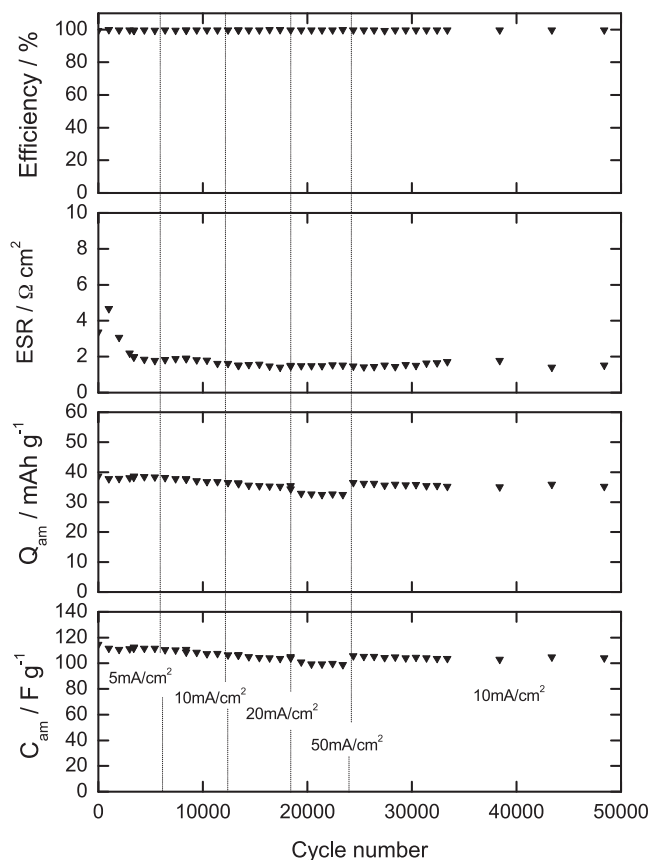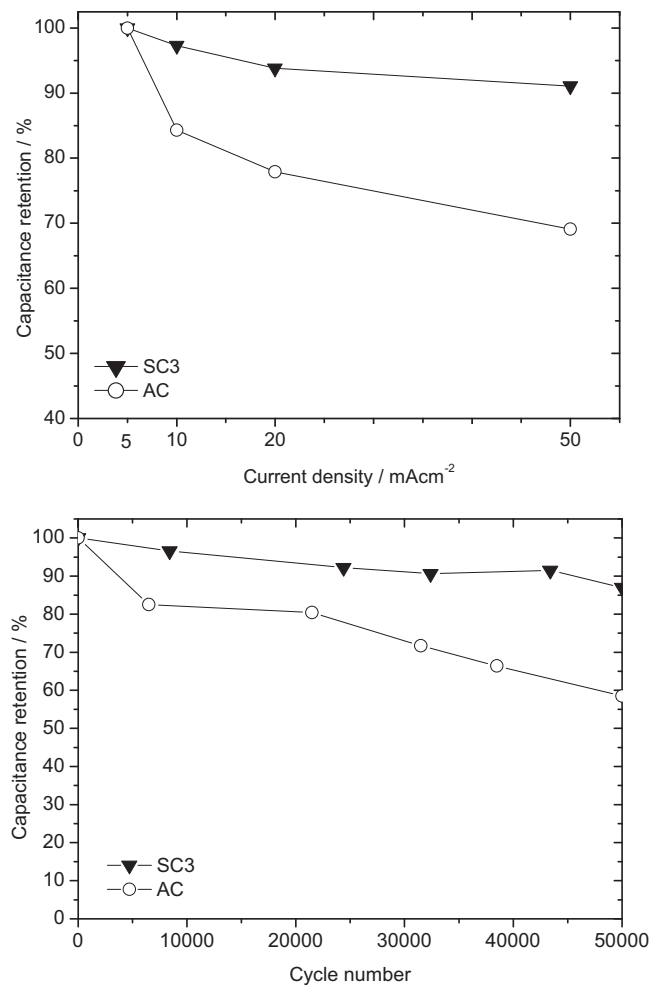
Fig. 4. Evolution of coulombic efficiency, ESR, specific capacitance (C_{am}) and specific capacity (Q_{am}) vs. cycle number for an EDLC based on SC3 carbon black obtained from galvanostatic charge–discharge measurements at 5, 10, 20 and 50 mA cm^{-2} in PC-Et$_4$NBF$_4$ (1 M).


Fig. 5. Comparison of capacitance retention of SC3 and DLC Super 30 electrodes during: (a) charge–discharge measurements at different current densities and (b) during prolonged charge–discharge tests (50,000 cycles) carried out in PC-Et$_4$NBF$_4$ (1 M).

specific capacity and capacitance around 32 mAh g^{-1} and 100 F g^{-1}, respectively.

The results of our comparative study of SC3 and DLC Super 30 are shown in Fig. 5. At the current density of 50 mA cm^{-2}, the SC3 electrode displays 91% of its initial capacitance and capacity (see Fig. 5a). In the case of AC-based EDLC the decrease was much higher and at 50 mA cm^{-2} the AC electrode displays only 70% of its initial capacitance and capacity (recorded at 5 mA cm^{-2}). As reported above, the SC3 carbon black particle size is smaller in comparison to that of activated carbon. Due to that, the length of pores within carbon black particles is shorter than that within activated carbons. Most likely, the presence of these short length pores (or, in other words, the absence of long pore lengths) plays a favourable role and it improves the electrode performance during the charge–discharge at high current. Furthermore, the SC3 electrodes display very good cycling stability. As shown in Fig. 5b, at the end of cycling (50,000 cycles), the capacitance retention for the SC3 electrodes is in excess of 90%. After the same numbers of cycles carried out under the same charge–discharge condition, the capacitance retention for the AC-based EDLC is only in the order of 60%.

Fig. 6 sums up the results from the galvanostatic charge–discharge experiments carried out with the two considered EDLCs in a Ragone plot of average energy versus power. The values on the plot are referred to the weight of active material (SC3) in two electrodes. Energy and power are calculated according to Eqs. (5) and (6), where i is the current in ampere (A), E is the voltage in volt (V), $m_{cell\text{-}active}$ is the active mass loading of the whole cell in grams (g) and t_d is the discharge time in seconds (s).
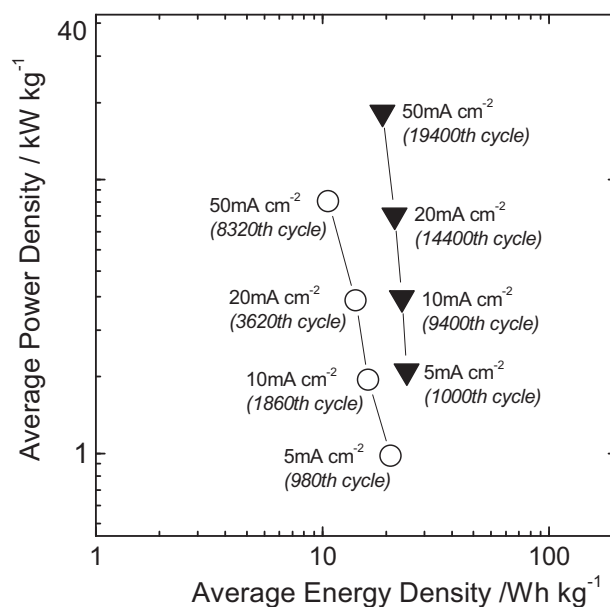
Fig. 6. Ragone plot for an EDLC based on SC3 carbon black. The average energy and power are referred to the total mass of active materials.

The integral refers to the discharge area between the highest and lowest voltage values.

$$E_{average} \ (Wh/kg) = i \times \int \frac{E}{m_{cell\text{-}active} \times 3.6} dt_d \tag{5}$$

$$P_{average}(kW/kg) = \frac{E_{average} \times 3.6}{t_d} \tag{6}$$

The energy and power values for the EDLC were taken at $5\,mA\,cm^{-2}$ (1000th cycle for SC3 and 980th cycle for AC), $10\,mA\,cm^{-2}$ (9400th cycle for SC3 and 1860th cycle for AC), $20\,mA\,cm^{-2}$ (14,400th cycle for SC3 and 3620th for AC), and $50\,mA\,cm^{-2}$ (19,400th cycle for SC3 and 8320th for AC). At $5\,mA\,cm^{-2}$, the SC3-based EDLC delivers an average energy and power of $23.5\,Wh\,kg^{-1}$ and $2\,kW\,kg^{-1}$, respectively. At $50\,mA\,cm^{-2}$, the SC3-based EDLC delivers an average energy and power of $18.4\,Wh\,kg^{-1}$ and $17.6\,kW\,kg^{-1}$, respectively. It is important to note that for all considered current densities, the values of energy and power obtained with the SC3-based EDLC were significantly higher than those obtained with the AC-based EDLC. In our opinion these values are useful to get information about the ability of SC3 to maintain good performance at relatively high currents, together with maintaining cycling stability. The cell shows a loss of only 8.5% in energy density after 24,400 cycles when the current was reduced from $50\,mA\,cm^{-2}$ back to $10\,mA\,cm^{-2}$.

In the past years, several studies investigated the performance of carbon blacks as active materials in EDLCs [18–23]. The studies indicated that carbon blacks are able to display high conductivity, high specific capacitance and have the advantage of improved pore accessibility. However, to the best of our knowledge, this report is the first example of a carbon black-based EDLC successfully tested for a large number of charge–discharge cycles in conditions similar to those normally applied for testing activated carbon-based EDLCs.

3.3. LIC based on SC3 carbon black

The interest in lithium-ion capacitors (LICs) has progressively been increasing in the last years. The combination of a lithium-ion battery electrode with an EDLC carbon-based electrode is a promising strategy for realization of devices able to display energy and power density values intermediate to those of lithium-ion batteries and EDLCs. LICs also have a high cycling stability superseding that of batteries. So far, several types of LICs have been proposed and investigated [13]. Among them, a device containing an EDLC carbon-based electrode as anode combined with a LiFePO$_4$-based electrode as cathode (C//LiFePO$_4$) is certainly of interest. In order to realize such a system, the selection of an anode with relatively high specific capacity appears to be of importance. As shown in the previous section dedicated to EDLC, SC3 carbon black, displaying a high specific capacity, high capacity retention at high current density and high cycling stability, appears to be one of the suitable candidates for LIC.

In order to evaluate the performance of SC3 as anode in LIC, SC3 electrodes were tested against metallic Li in our third series of experiments. CVs at $20\,mV\,s^{-1}$ were carried out in order to define the lowest operating potential for SC3 electrodes with the selected electrolyte (1 M LiPF$_6$/PC). Due to SC3 electrode serving as anode (negative electrode), we did not investigate the highest operating potential, instead, we used $2.8\,V$ vs. Li/Li$^+$ as such. We found that $1.25\,V$ vs. Li/Li$^+$ is the lowest limit of potential. This potential corresponds to one above which the coulombic efficiency of the charge–discharge process is higher than 98%. This value of potential is in good agreement with that observed by Wang et al. for an activated carbon used in combination with electrolytes containing PC as solvent and lithium salt [26]. In thus defined potential range (between $2.8\,V$ and $1.25\,V$ vs. Li/Li$^+$, corresponding to

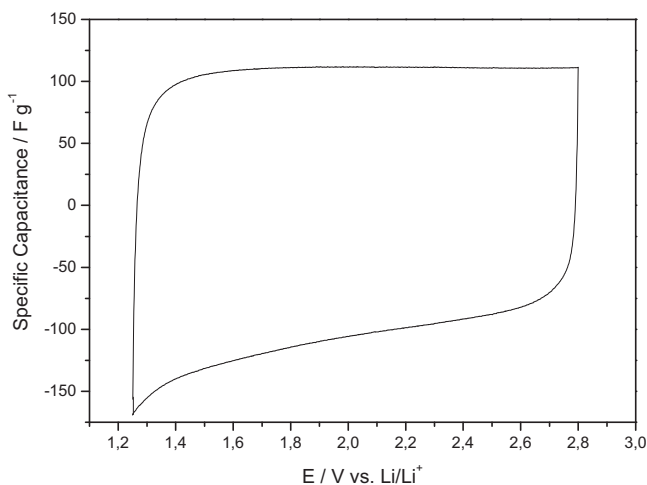**Fig. 7.** Specific capacitance values of SC3 carbon black electrode in PC-LiPF$_6$ (1 M) obtained from CV at $20\,mV\,s^{-1}$ by dividing the specific current by the scan rate.

$1.55\,V$), SC3 electrodes display a typical butterfly shape with a specific capacitance around $110\,F\,g^{-1}$, as shown in Fig. 7. In the same potential window, the charge–discharge tests at currents from 5 to $50\,mA\,cm^{-2}$ for a total of 12,000 cycles reveal the efficiency of the charge–discharge process of 100% (see Fig. 8). The initial ESR was in the order of $4\,\Omega\,cm^2$ and increased during the cycling process up to only $6\,\Omega\,cm^2$. The higher ESR of the system compared with the EDLC is most likely related to the lower conductivity of PC-LiPF$_6$ (1 M)

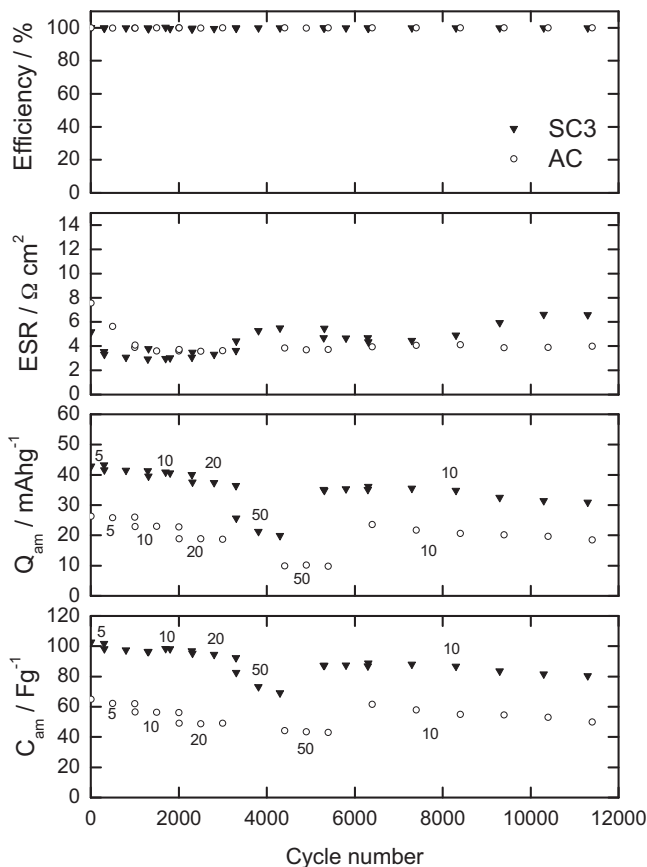**Fig. 8.** Evolution of coulombic efficiency, ESR, specific capacitance (C_{am}) and specific capacity (Q_{am}) vs. cycle number for a SC3 and DLC Super 30 electrode obtained from galvanostatic charge–discharge measurements at 5, 10, 20 and $50\,mA\,cm^{-2}$ in PC-LiPF$_6$ (1 M). Metallic lithium was used as counter electrode. The numbers correspond to the applied current density (e.g. 20 corresponds to $20\,mA\,cm^{-2}$).

compared with PC-Et$_4$NBF$_4$ (1 M) [27,28]. The constant increase of ESR during cycling is most likely related to the growing of an SEI layer on the lithium electrode. The effect is more pronounced at higher currents, promoting the formation of high surface area dendrites which react rather quickly with the electrolyte. At 5 mA cm^{-2}, the electrode displays a specific capacitance of about 100 F g^{-1}. However, at 50 mA cm^{-2}, the specific capacitance decreases to about 65–70 F g^{-1} with a continuous drop upon cycling. This is not observed at lower currents (5, 10 and 20 mA cm^{-2}), and could probably be related to the formation of SEI strongly pronounced at higher currents. At 5 mA cm^{-2}, the electrode displays a specific capacity of about 45 mAh g^{-1}. This value of capacity is slightly different compared to that shown by SC3 electrode in EDLC. The reason for this difference could be related to less conductive electrolyte and also different operating range of potential. Similarly to capacitance, the capacity decreases at higher currents. At 50 mA cm^{-2}, the electrode displays a capacity of about 20 mAh g^{-1}. It is nevertheless important to note that the electrode displays a very good performance in terms of cycling stability, specific capacitance and capacity for up to 12,000 cycles.

We also performed an analogous test for DLC Super 30 electrode against metallic Li. As shown in Fig. 8, the efficiency and cycling stability of AC electrode were comparable to that of SC3. The resistance of AC was initially about 8 Ω cm^2 and decreased during the test down to 4 Ω cm^2. This ESR of AC electrode seems to be less affected by the high current than the ESR of SC3 electrode, although both values are similar. Like in the case of SC3 electrode, both capacitance and capacity decrease at higher currents. However, it is important to note that the specific capacitance and capacity of AC were considerably lower than those displayed by SC3. At 5 mA cm^{-2}, the AC electrode displays a specific capacitance and capacity about 60 F g^{-1} and 25 mAh g^{-1}, respectively. These values are 40% lower than the values observed for SC3 electrode during the same test. At larger currents, the SC3 electrode also displays a capacitance and capacity higher than the AC electrode.

Based on the results of the tests against metallic Li, we concluded that SC3 may be an excellent candidate for a LIC with LiFePO$_4$ as cathode material. In this system, Li$^+$ ions move out from the LiFePO$_4$ electrode and a double layer is formed on the surface of SC3 electrode during charging process. During discharging, Li$^+$ ions move back from the electrolyte into the LiFePO$_4$ electrode, while the double layer on the SC3 electrode depletes. Therefore, we performed the fourth series of experiments using SC3 electrode against LiFePO$_4$ electrode. It is important to note that the LiFePO$_4$ electrode had a mass loading 3 times the mass of SC3 electrode. Indeed, our goal was to investigate the behaviour, and especially the cycling stability, of SC3 electrodes in a configuration where the source of Li$^+$ was a cathode based on LiFePO$_4$ and not a nearly infinite reservoir of Li$^+$ (e.g. metallic lithium). We used the same conditions as in the third series of experiments.

Fig. 9 shows the results of SC3 electrode against LiFePO$_4$ electrode in a LIC. The observed charge–discharge efficiency is around 100% for 12,000 cycles. The initial ESR is around 5 Ω cm^2 with a slight increase at the end of cycling. The specific capacity and capacitance at 5 mA cm^{-2} are comparable to those of SC3 against metallic Li, but, unlike to that system, the capacity and capacitance display stable values at higher currents (50 mA cm^{-2}) upon cycling. This difference could probably be related to the absence of dendrite growth and SEI upon cycling of LiFePO$_4$ electrodes at very high currents of 50 mA cm^{-2}. This observation and the ability of SC3 to maintain high capacity at high currents indicate a potential of this system for a powerful commercial LIC.

Finally, Fig. 10 shows the voltage profiles of SC3//LiFePO$_4$ system as well as SC3 negative electrode and LiFePO$_4$ positive electrode at current of 10 mA cm^{-2} (after 500 cycles). At this current, the LIC was able to deliver an average energy and power densities of
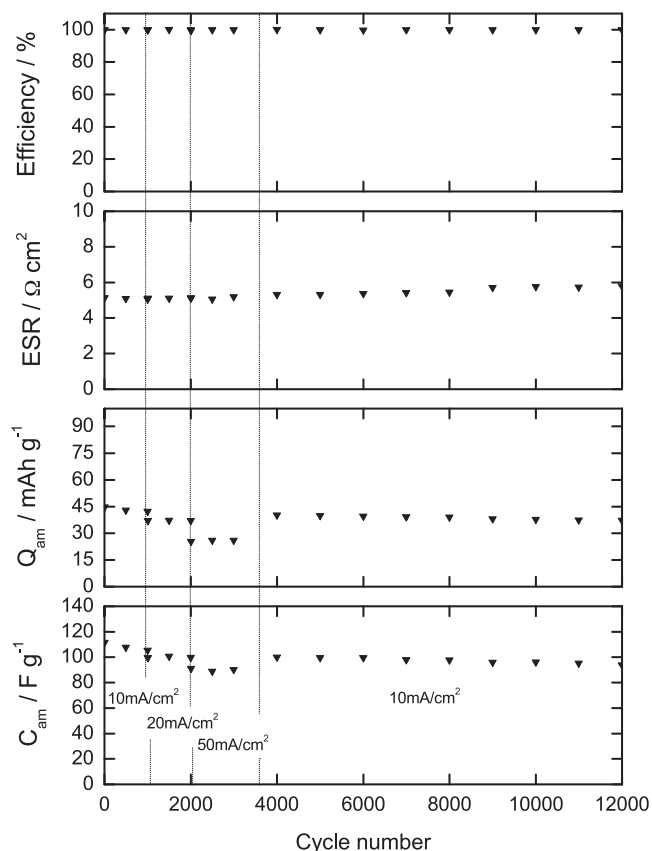

Fig. 9. Evolution of coulombic efficiency, ESR, specific capacitance (C_{am}) and specific capacity (Q_{am}) vs. cycle number for a SC3 electrode obtained from galvanostatic charge–discharge measurements at 5, 10, 20 and 50 mA cm^{-2} in PC-LiPF$_6$ (1 M). A LiFePO$_4$ electrode was used as counter electrode.

7.0 Wh kg^{-1} and 2.2 kW kg^{-1}, respectively, considering the weight of both active materials. These values are lower than those for SC3 EDLC, which is due to the fact that the electrodes in the LIC are not properly balanced. Only the SC3 electrode works in the desired voltage range. The LiFePO$_4$ electrode works in a very narrow potential window (approximately between 3 and 3.5 V vs. Li/Li$^+$). In our future work, a proper balancing of SC3/LiFePO$_4$ system will be necessary to improve the LIC performance, such as the energy density characteristics.
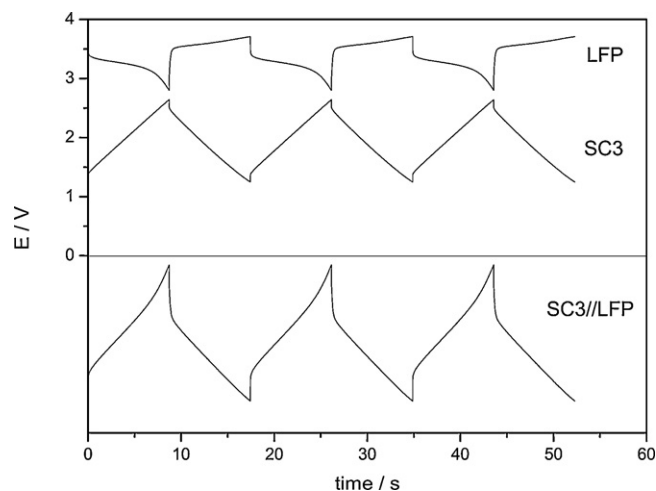

Fig. 10. (a) Voltage profile of a C//LiFePO$_4$ lithium-ion capacitor at 10 mA cm^{-2} in PC-LiPF$_6$ (1 M). (b) Voltage profile of a SC3 negative electrode. (c) Voltage profile of a LiFePO$_4$ positive electrode.

4. Conclusion

The SC3 carbon black from Cabot Corporation appears to be a promising active material for the realization of EDLCs and LICs.

SC3 in EDLC electrodes display a specific capacity and a capacitance of about $40\,mAh\,g^{-1}$ and $115\,F\,g^{-1}$ (both at $5\,mA\,cm^{-2}$), respectively. Very importantly, these values are not strongly affected at higher current densities. For instance, at $50\,mA\,cm^{-2}$, SC3 was able to display 91% of the capacitance and capacity obtained at $5\,mA\,cm^{-2}$. Moreover, the SC3-based EDLC displays very good cycling stability. After 50,000 cycles, SC3 displays capacitance retention of more than 90%. To the best of our knowledge, this report is the first example of a carbon black-based EDLC successfully tested for such high number of charge–discharge cycles in conditions similar to those normally applied for activated carbon-based EDLCs.

SC3 could also be used for the realization of a LIC, such as SC3//LiFePO$_4$. Here we showed an excellent cycling stability of this system for more than 12,000 cycles, together with the ability of the system to be charged–discharged with an efficiency of 100% at high currents of $50\,mA\,cm^{-2}$.

Acknowledgments

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