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Optimization of Carbon Electrodes for Solid-State E-Textile Supercapacitors

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Abstract. The integration of flexible supercapacitors (FSC) into electronic textiles (e-textiles) offers a promising power source, capable of fast charge-discharge rates, high-power density and long lifetimes. The design parameters for such a device are varied and complicated, with the need for low weight, cheap production, breathability and low toxicity. This work presents the optimization of the carbon electrodes within a solid-state, single-layer textile supercapacitor that meets these device requirements. Three commercial activated carbons are investigated with different ratios of activated carbon to Carbon Black, at differing loading levels. The maximum areal capacitance observed was 23.6 mF.cm⁻²

1. Introduction

Electronic textiles (e-textiles) add functionality to traditional fabrics through the integration of electronics into the structure, with interaction with the user and/or the environment. Energy harvesting, sensors and actuators can all be introduced into fabrics, enabling lightweight, flexible and miniaturised electronic circuits. The characteristics and design possibilities of e-textiles make them appealing to a myriad of sectors, from monitoring of physiological signals in Health [1] to embedded technology within the Commercial and Sport sectors [2]. However, e-textiles have been hampered by the choice of power source. Energy harvesting offers a source of power but is intermittent and battery packs offer a cumbersome addition to the fabric/garment. A promising solution is the integration of flexible supercapacitors (FSCs) into the fabric that, when combined with the energy harvesting, offer a reliable power source. Characteristics of fast charge-discharge rates, long cycle life and high power densities make FSCs favoured over batteries or traditional capacitor solutions.

FSCs can be in the form of parallel layers or yarn, each having advantages and disadvantages. Parallel layer FSCs suffer from encapsulation complications but improved contact resistance when scaled up. Yarn based devices have self-encapsulation during production but suffer from expensive manufacturing procedures. A solid-state, single-layer (SS-SL) fabric supercapacitor [3] offers a scalable, low cost solution that is competitive on capacitance with similar supercapacitors and can be seamlessly integrated into garments. In the SS-SL supercapacitor the two carbon electrodes, separator and electrolyte are all housed within the same fabric substrate. This work looks to optimize the carbon electrode for such a device.



2. Experimental

The experimental work within this study was split into two phases with the primary investigation involving the production of electrodes from three different commercial activated carbons with differing surface areas and pore dimensions. Along with looking at different carbons the number of layers sprayed onto the cotton was systematically investigated to try to optimize the whole system. Dealing with two discrete variables it was decided that a 3^k (where k is the number of variables, in this case 2) factorial experimental design was correct to capture the different influences. The second phase was to look at the ratio of Carbon Black to activated carbon. Carbon Black is added to the electrodes to help improve conductivity by better stacking of the carbon. This increases the electrical pathways within the porous structure but reduces the capacity of the electrodes due to a lower surface area than the activated carbon. The three commercial activated carbons used throughout this study were *Kuraray Chemicals YP-80F* (YP80F), *Cabot Norit GSX* (GSX) and *Cabot Norit SX Ultra* (SXU) with specific surface areas of 2200, 950 and 1200 $\text{m}^2\cdot\text{g}^{-1}$ respectively.

The electrode ink was produced by mixing 0.85 g of the carbon materials (in ratios 8:2, 9:1 and 1:0 activated carbon to Carbon Black (*Chevron Philips*)) with 0.15 g of ethylene-vinyl acetate dissolved (*Sigma Aldrich*) in 5 ml of 1,2,4-Trichlorobenzene (*Sigma Aldrich*). This ink was blended at 1000 rpm for three minutes in a *Hauschild Engineering* speed mixer before being further mixed in a *Branson 1510* sonic bath for 15 minutes. Once prepared, the ink was sprayed onto the cotton substrate. The cotton was held within an aluminum mask during spraying, which was then attached to a belt driven apparatus, figure 1. The spray gun was set at 25 psi and was 7.5 cm from the cotton for all of the devices. Each supercapacitor passed through the spraying arc for 0.5 s, and each pass was counted as one layer.

The electrolyte was prepared by dissolving 0.5 g of polyvinyl alcohol (*Sigma Aldrich*) in 5 ml of deionized water. 0.3 g of ammonium dihydrogen phosphate was then added and the electrolyte allowed to cool. The device was then constructed by submerging the electrodes in the electrolyte and placing the vessel under 50 mbar of vacuum for 20 minutes as the temperature was ramped to 40 °C. The electrodes were then removed from the electrolyte and compressed between two plates of acrylic and placed under 100 mbar of vacuum for 8 minutes at 40 °C. This vacuum impregnation technique was used to remove unwanted air from the electrode and increase the electrode wettability. The finished supercapacitor was then placed into a *Swagelok* cell for testing.

Electrochemical testing of the devices was performed with galvanostatic cycling (GC) at 0.25, 0.5, 1, 2, and 3 $\text{mA}\cdot\text{cm}^{-2}$ and cyclic voltammetry (CV) at 5, 25, 50, 100 and 200 $\text{mV}\cdot\text{s}^{-1}$ using a *Solartron 1470E Cell Test System*. The capacitance, power and energy densities were calculated from the GC measurements using equations (1), (2) and (3), where C is capacitance, E is energy, P is power, R_{ESR} is equivalent series resistance calculated from the instantaneous voltage drop and V_{peak} is the peak voltage. dV/dt is calculated from 80% to 40% of the peak voltage.

$$C=I \left(\frac{dV}{dt} \right)^{-1} \quad (1)$$

$$E=0.5CV_{\text{peak}}^2 \quad (2)$$

$$P= \frac{V_{\text{peak}}^2}{4R_{\text{ESR}}} \quad (3)$$

Scanning electron microscope (SEM) (*JEOL JSM-6500F*) images were used to supplement this testing.

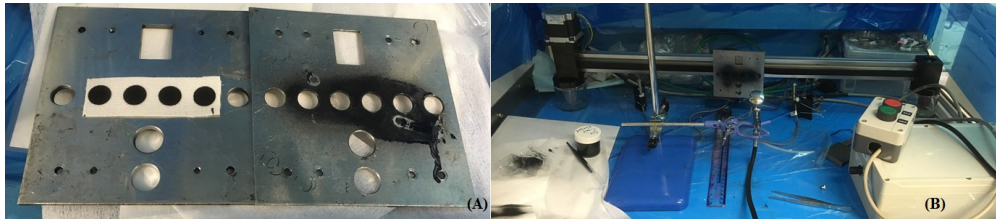


Figure 1. Aluminum mask (A) and spray rig (B) used for the production of the supercapacitors

3. Results and Discussion

SEM micrographs seen in figure 2 show an unsuccessful and a successful SS-SL supercapacitor. A clear, bright layer can be observed in the successful sample, signaling the manufacturer had been successful, with the carbon layers not permeating through the whole width of the substrate. This bright layer however, is not constant throughout the length of the device and could explain the instability of a number of the devices at higher current densities.

Table 1 presents the areal capacitance, power and energy densities for the devices characterized. The results presented are an average of five measurements on the best performing device of each batch. All of the devices produced with 6 layers of ink made from the YP80F activated carbon failed. This is attributed to a physical incompatibility of the ink to this process and could be due to an uneven distribution of the carbon within the ink. This is supported by the results seen in Table 2, where again a high loading of YP80F failed to produce a working device. The highest recorded areal capacitance was 23.6 mF.cm^{-2} which is comparable, or better, than devices produced from more complex (and costly) materials and techniques (Zang *et al* 23 mF.cm^{-2} [4], Abdelkader *et al* 2.5 mF.cm^{-2} [5]). A more detailed investigation of the highest performing device can be seen in figures 3-5. The CV profile (figure 5) at lower sweep rates is rectangular, showing its low losses. At higher sweep rates the symmetry of the profile remains, showing the stability of the device under harsher operating conditions. The degradation (61% loss between 0.25 and 3 mA.cm^{-2}) in areal capacitance, seen in figure 4, shows that further work is required if these devices are to be used for high power applications.

Table 2 shows the results of tests where the ratio of Carbon Black to activated carbon was changed. The highest capacitance recorded in these test was 10.9 mF.cm^{-2} with the configuration of 80% YP80F activated carbon to 20% Carbon Black, with 6 spraying layers. This was the only configuration of 6 layers of YP80F based ink to produce a working device. This is attributed to a more even dispersion of carbon through the ink and better conductivity of the electrode. These tests demonstrate that though the Carbon Black has advantages however, for an increase capacitance, this must be replaced as much as possible.

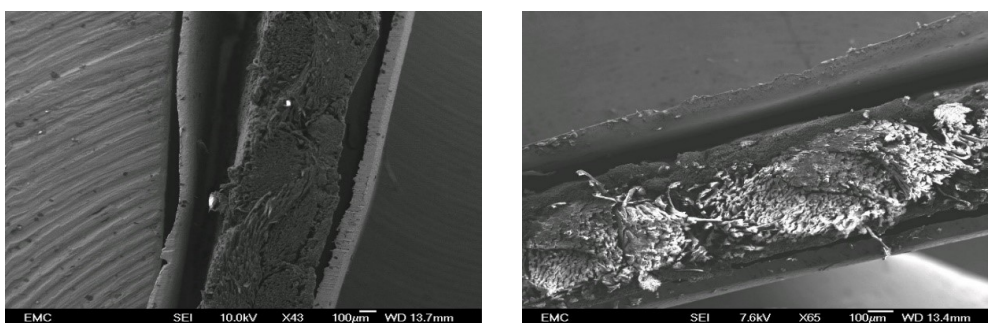


Figure 2. An SEM comparison of an unsuccessful (left) and successful (right) supercapacitor

Table 1. Comparison of the performance of the SS-SL supercapacitors at a 90% activated carbon ratio.

Device	No. of Layers	Performance Characteristics		
		Areal Capacitance (mF.cm ⁻²)	Energy Density (μWh.cm ⁻²)	Power Density (x10 ⁻⁴ W.cm ⁻²)
YP80F	2	2.7	2.4	2.0
YP80F	4	23.6	2.1	5.9
YP80F	6	-	-	-
SXU	2	1.5	0.13	4.6
SXU	4	1.2	0.10	4.3
SXU	6	1.1	0.10	5.3
GSX	2	1.5	0.13	4.6
GSX	4	1.2	0.10	2.8
GSX	6	1.2	0.11	4.1

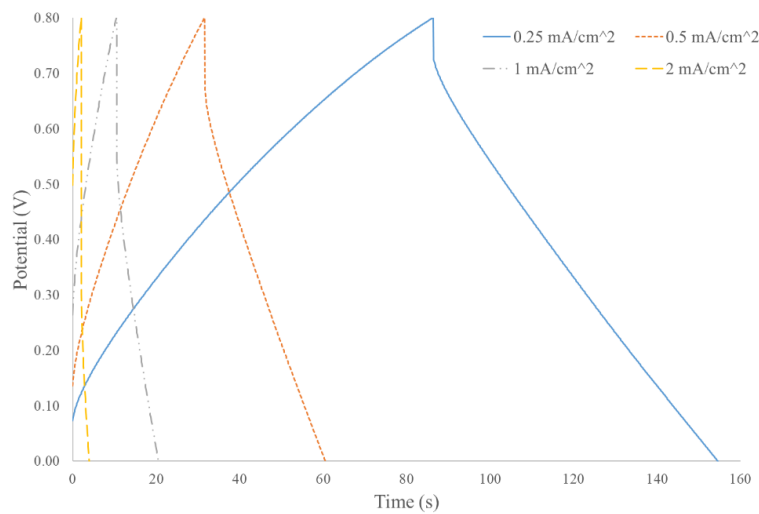


Figure 3. GC measurements for the 4 layered 90% YP80F supercapacitor

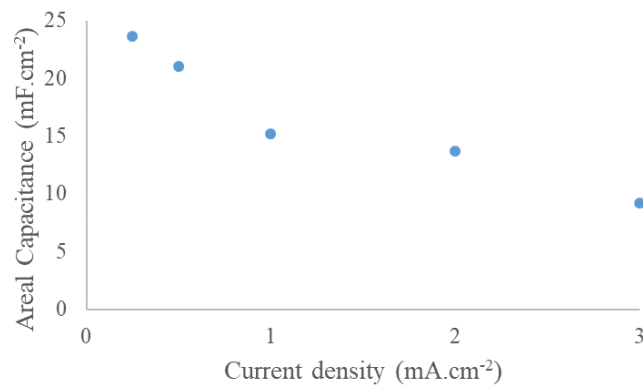


Figure 4. Capacitance degradation with increasing current density

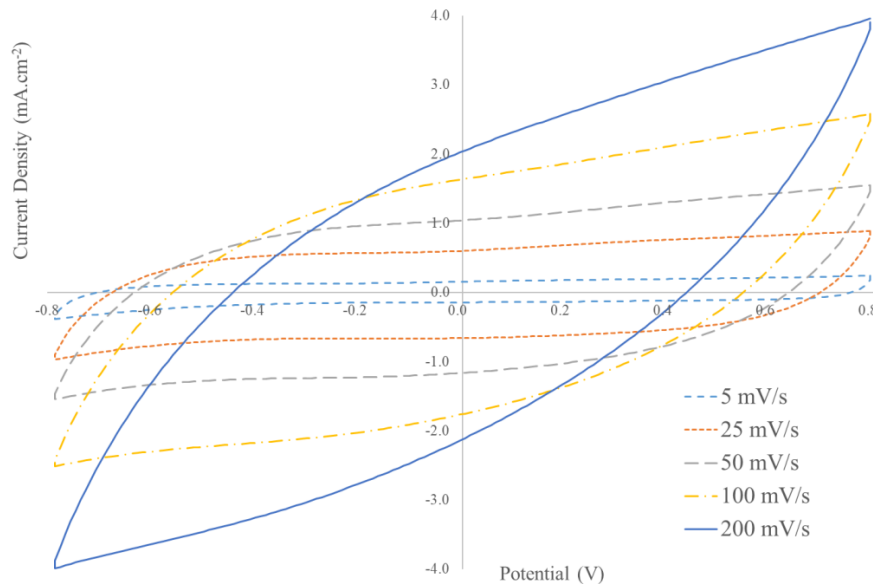


Figure 5. CV plots from 5 to 200 mV/s for the 4 layered 90% YP80F device

Table 2. Comparison of devices produced with varying ratios of Carbon Black

No. of Layers	Device	% of Activated Carbon	Performance Characteristics		
			Areal Capacitance (mF.cm ⁻²)	Energy Density (μWh.cm ⁻²)	Power Density (x10 ⁻⁴ W.cm ⁻²)
4		80	0.26	0.02	1.1
6		80	10.9	0.97	3.3
4		100	-	-	-
6		100	-	-	-

4. Conclusion

This work has presented a study to optimize the performance of a SS-SL textile supercapacitor. Three different commercial activated carbons were investigated in 13 configurations. A maximum areal capacitance of 23.6 mF.cm⁻² was observed. These devices offer an affordable and scalable solution to the power requirements of modern e-textiles.

5. References

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