Upscalable ultra thick rayon carbon felt based hybrid organic-inorganic electrodes for high energy density supercapacitors

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Abstract
Low weight, small footprint, and high performances are essential requisites for the implementation of energy storage devices within consumer electronics. One way to achieve these goals is to increase the thickness of the active material layer. In this work, carbonized and graphitized rayon felt, a cellulose-derived material, is used as a three-dimensional current collector scaffold to enable the incorporation of large amount of active energy storage materials and ionic liquid-based gel electrolyte in the supercapacitor devices. PEDOT:PSS, alone or in combination with active carbon, has been used as the active material. Three-dimensional supercapacitors with high per unit area capacitance (more than 1.1 F/cm²) have been achieved owing to the loading of large amount of active material in the felt matrix. Areal energy density of more than 101 μWh/cm² and areal power density of more than 5.9 mW/cm² have been achieved for 0.8 V operating voltage at a current density of 1 mA/cm². A nanographite material was found to be beneficial in reducing the internal serial resistance of the supercapacitor to lower than 1.7 Ω. Furthermore, it was shown that even after 2000 times cycling test, the devices could still retain its performance with at least 88% coulombic efficiency for all the devices. All the materials are readily available commercially, environmentally sustainable and the process can potentially be upscaled with industrial process.

KEYWORDS
active carbon, organic-inorganic hybrid supercapacitors, PEDOT:PSS, rayon carbon felt, ultra thick electrodes

INTRODUCTION

Supercapacitors, an important energy storage device that facilitates the digitalization, electrification, and sustainability of the society, are finding increasing usage in a variety of applications ranging from electric vehicles to consumer electronics.1,2 In recent years, the continuous advancements in wearable electronics boosted the...
Tubular MnOOH has been grown on carbon felt using the hot filament lasering process,\textsuperscript{14} showing promising hierarchical structure that benefits capacitance retention.\textsuperscript{25} Polypyrrole/MnO\textsubscript{2} combination has also been tested.\textsuperscript{26} Tubular MnOOH has been grown on graphite carbon felt.\textsuperscript{27}

Another explored approach consists of the impregnation of the carbon felts with conductive materials such as, for example, CNT/graphene\textsuperscript{28} and graphene/MnO\textsubscript{2}.\textsuperscript{29}

However, the often complex and costly processes, and expensive raw materials such as Mn and Co are not ideal candidates for sustainable energy storage/supply solutions with the demand for more environmentally friendly process and material combinations rising.

As a non-metal containing alternative, organic conducting polymer materials, for example, polypyrrole (PPY), poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS; here after referred to as PEDOT), have been increasingly used for supercapacitor applications. Amid possessing advantages to be solution processable, they hold the promises to flexible devices and ease for recycling.\textsuperscript{9,11,30}

Taking the above factors into account, in this work, we have devised thick 3-D hybrid electrodes for high areal density supercapacitor application by combining rayon carbon felt and “green” active materials. Rayon carbon felt acts as both, a scaffold to retain active materials and a 3-D current collector thanks to its relatively good electrical conductivity. Commercially available materials such as PEDOT or a mixture of PEDOT with activated carbon (AC), have been incorporated, as active materials, inside the felts 3-D conducting felt fiber matrix. Nanographite material has also been tested as a way to further reduce the internal resistance of the device. Once the carbon felt was electrochemically activated, the impregnation of active material was performed by combining drop casing and cold pressing methods. Loadings of up to 1 mm into the bulk of the felt has been realized for each active material combination, which demonstrates an excellent route to achieve high energy density storage devices. We also show that an ionic liquid gel can be effectively implemented and held in the felt preloaded with active materials. For supercapacitors fabricated using a PEDOT and AC mixture as the active material, high areal capacitance up to 1.1 F/cm\textsuperscript{2} and relatively low ESR of approximate 2 \textOmega \textsuperscript{2} were recorded. The route presented herein suggests a promising fabrication approach to achieve ultra thick 3-D supercapacitor with high energy density that also has potential in batteries technology. To the best of our knowledge, this is the first time Rayon carbon felt is used for 3-D ultra thick supercapacitor applications.
2 | EXPERIMENTAL

2.1 | Ink preparation

Carbonized and graphitized carbon felt Sigracell GFA 6EA based on rayon was used for this investigation. The felt has a BET surface area of 0.8 m²/g and an open porosity of 95%. The felt was 6 mm thick originally but it was thinned down to 3 mm by using a scalpel to shear through the felt. PEDOT:PSS used in this work was Orgacon DRY Pellets purchased from AGFA. Activated carbon, using coconut shell as the starting material, was supplied by Eurocarb. Clean room wipe was used as a separator material for the supercapacitor. Nanographite (NG) with trade name of xGnP was purchased from XG Sciences. Ethylene glycol and glycerol were both acquired from Sigma-Aldrich. Gel electrolyte was prepared by using ionic liquid material 1-ethyl-3-methylimidazolium ethyl sulfate (EMIM-ES) with the addition, as a gelation agent, of 5% (of total mixture weight) of 2-hydroxyethyl cellulose (2-HEC; Mv/C24 90 000) from Merck (Sigma-Aldrich 534 965). The electrolyte mixture was prepared by dissolving 2-HEC in EMIM-ES at around 75°C.

The different active inks formulation evaluated in this work are shown in Table 1. The mixtures, named as PEDOT, AC/PEDOT, and NG/AC/PEDOT inks, were homogenized by a speed blender (DAC 600.1-CM 50). The mixing speed was 8000 rpm and the duration was 8 min.

2.2 | Manufacturing of supercapacitors

The process flow for the supercapacitors assembling is summarized in Figure 1.

First, in order to make the carbon felt surface more hydrophilic and improve impregnation of the active materials, electrochemical surface treatment was performed. This treatment was achieved by applying anodic pulses of 1.8 V for 300 seconds followed by −1.5 V for 60 seconds in a phosphate buffer solution for 3 cycles. The aqueous buffer solution used in this pretreatment step is composed of 0.05 mol/L phosphate buffer solution (pH 7.0) containing 500 mmol/L of Mg(NO₃)₂. A three-electrode setup consisting of a Ag/AgCl (3 mol/L NaCl) reference electrode, a stainless steel mesh as a counter electrode, and the carbon felt as a working electrode was used. Electrochemical measurements were performed using an Ivium Octostat potentiostat/galvanostat. Afterward, the felts were thoroughly washed with DI water and dried in the oven at 110°C.

Next, the felt was attached to a flexible Al foil with PET backing (current collector) using a thin layer of printable carbon ink as a conductive adhesive in-between layer. The 9-μm Al/36-μm PET laminated foil was from Skultuna induflex. Dupont Carbon 7102 ink was used as both the conductive adhesive material and the protective layer for the Al current collector. The carbon ink was printed on top of Al by stencil printing; following assembling the structure was thermally cured at 90°C for 15 minutes.

Afterward, the active material ink was drop cast and cold pressed into the felt using a manual pressing tool. Following impregnation, the wet felt electrodes were first dried inside an oven at 90°C for 2 hours and then cured at 120°C for half an hour.

Three types of electrodes were fabricated using different kinds of active materials listed in Appendix S1, Table I, that is, PEDOT, AC/PEDOT, and NG/AC/PEDOT, respectively. All the three kinds of impregnated electrodes have felt areas of 4 × 4 cm². As a reference, some electrodes were prepared also using only surface-treated felts (no active materials).

Finally, following the impregnation of the electrolyte in the felt and in the paper separator, the supercapacitors were assembled by aligning two electrodes (of same ink composition) and the separator vertically.

2.3 | Characterization

To characterize the wettability of the developed inks on the carbon felts, contact angle measurements were performed. The inks were dropped on the felt surfaces and
the angles were captured from the side. The goniometer instrument used in these measurements was from Ramé-hart.

The electrode morphology was studied by a scanning electron microscope (SEM) using a Zeiss Gemini (model Sigma 500) SEM system. Both planer view and cross sections were investigated. The cross section of the sample was prepared by cutting the sample straightly vertically using a scalpel.

Supercapacitor performance was evaluated using an Ivium Octostat. Cyclic voltammetry (CV), DC current charge-discharge, and charge-discharge cycling measurements have been done. Different supercapacitor performance parameters were extracted in accordance with the procedures described by Zhang et al. Electrochemical impedance spectroscopic measurements of the supercapacitors were performed with a Biologic potentiostat SP-200 using an amplitude of 7 mV in the frequency range of 10 mHz to 100 KHz.

A demonstrator was made by driving an electrical fan using two supercapacitors in serial connection.

## 3 RESULTS AND DISCUSSIONS

Three device configurations have been investigated to demonstrate the proposed ultra thick green electrodes concept and by using three sets of inks (Table 1), respectively. For the carbon felt supercapacitor based only on PEDOT as the active material, the pressing onto/into the treated felt was relatively easy. However, due to the high-water content of the PEDOT ink, it was not possible to load a large amount of PEDOT materials unless multiple repetitions of the loading and drying processes were performed. The high amount of water in the PEDOT ink was, however, necessary in order to achieve adequate viscosity for the ink to penetrate into the felt and adhere to the carbon fiber surface. AC/PEDOT and NG/AC/PEDOT inks had much higher solid content from the beginning, and subsequently, it was possible to load higher amount of active materials with only a single round of loading and drying. PEDOT was proven to be critical for the adhesion of the AC and NG/AC to the treated rayon carbon felt, as shown by the contact angle measurements (see Appendix S1, SI-1 section). Without PEDOT, it was very difficult for AC or NG/AC water suspensions to penetrate into and remain inside the treated carbon felt. The results of the loading experiments seem to indicate that PEDOT is a key ingredient on the active material ink formulations serving as an electroactive wetting/adhesion promoting component. The satisfactory mixability between PEDOT and different sorts of carbonaceous materials is attributed to the different kinds of bonds such as C-S, C-O-C, and C-C that can be present, and enhanced π-π binding.

The original felt structure contains large portion of voids (ca. 95%), which are well suited for filling in with additional active materials. Appendix S1, SI-2 section presents SEM images of the carbon felt without active materials loaded; clearly voids of several tens of μm can be seen. For the felts loaded with active materials but without gel electrolyte, SEM imaging provided insight about the surface coverage and vertical direction penetration of the ink into the felt. Figure 2 shows the plan view and the cross-sectional images of the samples with active materials. It can be observed that the felt surfaces (Figure 2A,C and E) are mostly covered by the active materials. The cross-sectional images (Figure 2B,D and F) show that the inks have penetrated inside the felt structures; considerable amount of active materials can
be seen for all three kinds of inks (Table 1) up to ca. 1 mm in depth. The images also show that even though there is a high loading of the active material into and onto the felt, there is still quite much porosity remaining, leaving room for the electrolyte that will be pressed into the felt in the following step.

Two electrode electrochemical measurements were performed on the assembled supercapacitor devices. All the three assembled devices were, independently from having PEDOT, AC/PEDOT, and NG/AC/PEDOT as active materials into the felts, nearly symmetrical, considering the geometry and the amount of active materials and gel electrolyte loaded inside. Figure 3 shows CV, charge-discharge, and impedance measurements for the three types of supercapacitors, respectively. CV measurements (Figure 3A and B) show that the supercapacitor based solely on PEDOT behaves closely to ideally capacitive for the voltage range (0-0.8 V). The non-pseudocapacitive behavior of PEDOT has been observed previously and it has been ascribed to the double-layer forming around the PEDOT chains. AC/PEDOT- and NG/AC/PEDOT-based supercapacitors exhibit both much larger capacitances and non-ideal capacitive curves that could stem from the different charging mechanisms and kinetics between active carbon and the electrolyte. If at low scan rates the supercapacitors with the different active materials follow with the scan duly, for higher scan rates the AC/PEDOT- and NG/AC/PEDOT-based supercapacitors show unproportionally lower current due to the large mass and the slower kinetic involved. CV
measurements at higher scan rates are shown in Appendix S1, SI-3 section.

Figure 3C presents the discharging profiles for charge-discharge measurements on the supercapacitors. All the three active material combinations have quite large capacitances with areal capacitance (at the current density of 1 mA/cm²) for PEDOT, PEDOT/AC, and NG/AC/PEDOT, respectively, of 0.2, 1.14, and 1.16 F/cm². Compared to the three supercapacitors loaded with the different active material combinations (Table 1), the reference device (carbon felt no active material) showed a remarkably lower areal capacitance of 2.63 mF/cm² and thus its contribution to the capacitance is negligible. This result also confirms that the surface treatment only has made its surface more hydrophilic. In Appendix S1, Figure SI-4, the charge-discharge measurement on the reference device is presented. The NG has a rather limited (a few F/g) charge storage capacity, so its contribution for capacitance of the device is also ignorable. Subsequently, the higher capacitance of the AC/PEDOT and NG/AC/PEDOT supercapacitors, when compared to the supercapacitor containing only PEDOT, is thus directly related to the loading of the active carbon materials inside the rayon carbon felt. From charge-discharge measurements, the ESR values were derived for PEDOT, AC/PEDOT, and NG/AC/PEDOT supercapacitor devices to be 2.94, 2.38, and 1.69 Ω, respectively. The lower ESR for the AC/PEDOT supercapacitor compared to PEDOT only supercapacitor is attributed to high amount of filling of AC materials. The presence of NG, thanks to its remarkable conductivity, further decreases the ESR. Impedance measurements (Figure 3D) confirmed that these devices have relatively low ESR values amid the relatively high thicknesses of the active materials and carbon felt composite compared to conventional supercapacitor constructions. Impedance fittings is presented in Appendix S1, SI-5 section. A summary of the characteristics extracted from the charge-discharge and other electrochemical measurements for the supercapacitor devices is presented in Table 2. As it can be derived from Table 2, already at 0.8 V and 16 mA operational conditions, the developed
supercapacitors are well positioned in the areal-normalized Ragone plots, as represented by the areal energy density of 17.8, 101.1, and 103.1 μWh/cm²; and areal power density of 3.40, 4.21, and 5.93 mW/cm², for PEDOT, AC/PEDOT, and NG/AC/PEDOT supercapacitors, respectively. It should be noted that for AC/PEDOT and NG/AC/PEDOT supercapacitors, the working voltage range could be further extended toward higher voltages with the ionic liquid gel electrolyte used.

In Appendix S1, SI-3, more electrochemical measurements data are provided, along with CV scans at different scan rates or broader voltage range, charge-discharge measurements at different current levels or with higher voltage limits are also shown.

To investigate the robustness of the developed supercapacitors, 2000 cycles of charge-discharge measurements were performed (Figure 4). Satisfactory results have been achieved in terms of retention of both

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**TABLE 2** Extracted values for supercapacitors at 16 mA and 0.8 V; area for each supercapacitor is 4 × 4 cm²

|                     | PEDOTsupercap | AC/PEDOTsupercap | NG/AC/PEDOT supercap |
|---------------------|---------------|------------------|----------------------|
| Active material (PEDOT and AC, excluding PSS and NG) in device (g) | 0.181 | 1.663 | 1.752 |
| Capacitance (F) for supercapacitor device | 3.2 | 18.19 | 18.55 |
| ESR (ohm) for device | 2.94 | 2.38 | 1.69 |
| F/g (PEDOT and AC, excluding PSS and NG) for electrode | 70.72 | 43.76 | 42.35 |
| P (W/Kg) for device | 300.90 | 40.52 | 54.11 |
| E (Wh/Kg) for device | 1.57 | 0.973 | 0.942 |
| Areal capacitance for device (F/cm²) | 0.2 | 1.14 | 1.16 |
| E (μWh/cm²) for device | 17.8 | 101.1 | 103.1 |
| P (mW/cm²) for device | 3.40 | 4.21 | 5.93 |

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**FIGURE 4** Cycling measurement results. A, columbic efficiency; (B) capacitance retention; cycling was done at 9 mA and to 0.6 V for PEDOT; for AC/PEDOT and NG/AC/PEDOT, 160 mA and 1.4 V were used, respectively. Columbic efficiency is defined as the ratio of discharge time vs charge time for every cycle; and capacitance retention is defined as the ratio between capacitance for every cycle with respect to the initial capacitance value from the first cycle.

**FIGURE 5** Demonstrator. Each of the two supercapacitors has a size of 2 × 2 cm². A, picture for the set-up; (B) circuit schematic.
capacitance and Columbic efficiency. It should be noted that different current levels and voltage levels were used for different supercapacitors to reflect the different working voltage range and capacitance values the supercapacitors possess. After 2000 cycles, PEDOT showed good Columbic efficiency (>97%) but the capacitance retention ratio dropped gradually with the elapsed cycles to around 78% of the starting value at the end of the test, indicating degradation of PEDOT with repeated usage.\textsuperscript{36} AC/PEDOT and NG/AC/PEDOT showed lower Columbic efficiency compared to PEDOT, with values >90% and 88%, respectively. Both AC/PEDOT and NG/AC/PEDOT showed better capacitance retention, with values of >100.4% and 93%, respectively. Overall, the presence of the PEDOT, essential to enable the loading on the active materials, into the AC/AC and NG/AC/PEDOT supercapacitors seems not to have a negative impact on the performance of the AC/AC and NG/AC/PEDOT supercapacitors.

The developed supercapacitors are presenting characteristics well suited for powering up a range of sensor, actuator, and communications systems, and due to their flexibility, they are also suitable for wearable devices.\textsuperscript{3,37} To show their capability, two supercapacitors in serial were used to drive an electric motor (Figure 5 & Video S1).

4 CONCLUSIONS

To summarize, the high areal capacitance and the relatively low ESR obtained for the different supercapacitor devices prove that the combination of carbon felts as a 3-D conductive scaffold with PEDOT and carboneous active materials can be a promising approach toward the manufacturing of ultra thick supercapacitors. The results of this investigation indicate the importance of the active ink formulation to ensure its good penetration in the felt (eg, presence of the PEDOT for active carbon) and good performances (eg, presence of AC for overall capacitance and NG for low ESR). The obtained supercapacitors showed state of the art performance with areal capacitance larger than 1.1 F/cm\textsuperscript{2}, areal energy density above 101 μWh/cm\textsuperscript{2}, and power density of higher 5.9 mW/cm\textsuperscript{2} for 0.8 V operating voltage at a current density of 1 mA/cm\textsuperscript{2}. Furthermore, 2000 times cycling tests showed no severe degradation in the performances of the supercapacitors.

It must be noticed that in this proof-of-concept work penetration of active materials of ca. 1 mm into the carbon felt was obtained; this seems to leave room for further improvement in the device in terms of process and material optimization. This could be achieved by, for example, moving from a manual process to an instrumental process (eg, by using industrial impregnation processes) or by further adjusting the material composition (eg, ink formulation) or materials selection (eg, structure of felt). The better filling of the felt will further increase the device capacity and decrease the ESR for the supercapacitor.

Finally, since all the materials are commercially available in large amounts, our process can be expected to be easily transferable to industrial upscaling.

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DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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