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Chapter

Fabric-Integrated, Ionic Liquid-Based Supercapacitor as a Tunable and Flexible Power Source

Sathya Narayan Kanakaraj, Paa Kwasi Adusei, Yu-Yun Hsieh, Yanbo Fang, Noe Alvarez and Vesselin Shanov

Abstract

With the introduction of flexible and wearable electronic technologies such as displays, antenna's, etc., there has been an increased need for integrable, easily scalable, and safe electric power sources. Advances in flexible lithium-ion batteries have been recently reported, however they may still suffer from potential thermal runaways. In this chapter we review the progress in the topic of wearable energy storage devices. These devices have taken the form of both sheets and fibers entirely made of active material. We also discuss the advantages and drawbacks of each forms. Finally, we present our own work revealing a simplistic way to integrate working carbon electrode materials into suitable textile and to functionalize the obtained flexible structure with ionic liquid thus creating fabric supercapacitors. These devices can then be connected easily in series (9 V) or in parallel (high current), depending on the current or voltage requirements. The area of the electrodes can also be tuned to sustain higher capacitances. We report an energy density of 48 Wh/kg for a functional device at 3 V working window, which reveals no losses in energy density after 10,000 bending cycles.

Keywords: supercapacitor, $EMIMBF_4$, wearable electronics, gel electrolyte, energy storage

1. Introduction

Wearable technology has seen a great spike in development over the past decade [1–3] in the form of fabric integrated sensors (heart rate, chemical gas, etc.), information transfer lines and even energy harvesting (piezoelectric). This comes with the need to develop flexible and durable devices that can effectively power them [4, 5]. Energy storage devices typically consist of current collectors (for transfer of current to and from the electrodes), high surface area electrodes (for actively storing energy in the form of ions), insulating separators (for preventing shorting between electrodes) and an electrolyte. High surface area carbon such as graphite and activated carbon has become the standard electrode for many mainstream energy storage applications. However, metals (copper, steel and aluminum) are still heavily relied upon for taking on the role of current collector. This is a major issue for wearable devices as metals very easily succumb to fatigue through bend cycles and are also prone to oxidation. In addition, metal electrodes substantially add to

the total weight of the device. Care must be taken to prevent the metal from having direct contact with electrolyte as this would cause unwanted oxidation reactions and subsequent deterioration of the device. All these factors combined make it necessary to find an alternative to the metal current collectors for developing wearable energy storage devices.

Carbon based nanomaterials have emerged as promising candidates for the role of a flexible and durable current collector [6]. They boast very high theoretical specific surface area [7–11] reaching up to 3100 $m^2 g^{-1}$ for graphene [11]. Some of the materials also have very high strength and conductivities owing to their sp^2 bonds [12]. All these properties are valuable when it comes to energy storage. Further, most of carbon-based nanomaterials can be synthesized employing non-expensive and scalable processes, making them an ideal alternative to metals. These materials can even be tuned and equipped to function differently depending on the application and need (energy vs. power) represented in the Ragone plot in **Figure 1a** [13].

Electric double layer capacitors (*EDLC*) are employed where the power is of utmost importance, i.e. the ability to charge and discharge quickly. This is achieved through a non-reactive mechanism wherein the ions of the electrolyte are simply housed on and in the electrodes [14–17]. This process is depicted in **Figure 1b**, the pore size and volume being the most important parameters affecting the energy stored. The size of the pores must be very similar to that of the ion size, meaning different electrolytes require different pore size distributions. Since the mechanism of ion housing is a simple potential assisted diffusion, it allows for very high power [18–20] and high stable cycling of over 100,000 cycles. However, the stored energy is limited as it is heavily restricted by the achievable pore density. This can be partially overcome by using higher voltage stable electrolytes. Ionic liquids are one such class of electrolytes that have a stable window of about 4.5 V.

Pseudocapacitors are a class of energy storage devices that fit the need for high power without compromising on energy. The electrode materials used here have the disadvantage of being low in conductivity, leading to heavy losses through resistance. This is overcome by depositing them on conducting templates. They rely on fast redox reaction mechanism, usually involving surface absorption of electrolytic cations and proton absorption that leads to change in oxidative state [13, 21]. This involves protonation/electronation of the electrode resulting in a change in oxidation state. The briefly described mechanism is depicted in **Figure 1c**. Since redox reactions require actual chemical change, the electrode unavoidably degrades over time due to inefficiencies. The degradation is also attributed to volume expansions that cannot be sustained by the poor mechanical properties of the material. This results in lowering of the cyclability considerably [22]. Another consequence of the higher energy is lowering of the power. To fully utilize the electrode surface area, engineered nanostructures of pseudocapacitive materials with high surface area to volume fractions needs to be realized.
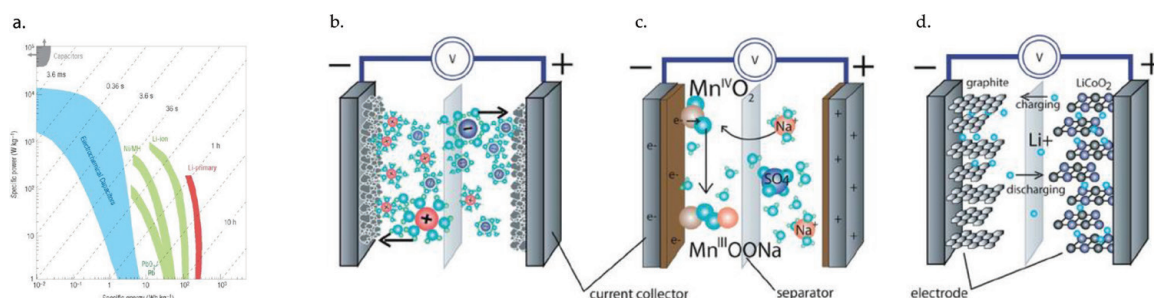
Figure 1.
(a) Ragone plot of specific power vs. specific energy for different energy storing devices—reproduced from [13] with permission from Springer Nature; (b) EDLC; (c) pseudocapacitors; and (d) battery—reproduced from [17] with permission from The Royal Society of Chemistry.

Batteries are electrochemical devices that fulfill the needs of high energy storage densities at the expense of power reduction. Their charge-storing mechanism involves actual chemical reactions taking place at the electrodes [23]. This includes ions reacting and transferring from one electrode via the electrolyte and intercalating into the other electrode, as displayed in **Figure 1d**. Depending on the chemical makeup of the electrode and the electrolyte used, the voltage window and charge stored can be tuned [24]. Due to the formation of actual compounds, the reversibility of this mode of charge storage is limited. It also gives the lowest power density of all the energy storage mechanisms described here.

All the mechanisms discussed above can be taken advantage of by using carbon-based nanomaterials to make them adapt to a wearable form. Energy storage in the form of wearable devices has seen considerable development in recent years and has achieved 3 major milestones (methods).

(i) Introduction of sp^2 bonded carbon nanomaterials.

(ii) Employment of dip coating and its optimization.

(iii) Synthesis of fiber-based devices.

The intention of this chapter is to give a concise view of the progress that the wearable energy storage research has achieved, to discuss the advantages and drawbacks of each milestone and finally to introduce our approach for fabricating integrated energy storage devices.

2. Choosing electrode material

Conventional fabric materials do not inherently have the ability to store energy. Thus, the materials that functionalize and allow textile fabric to become energy storage devices are of high importance. It is necessary to explore and develop these materials, mainly carbonaceous, to best suit the application. Owing to this, there has been extensive research conducted on the synthesis/fabrication and characterization of carbon nanomaterials.

The most simplistic form of carbon nanomaterials is the exfoliation of coal to create *carbon black and activated carbon* [25, 26] (**Figure 2a**). The latter has a very high porosity in both the micro (<2 nm) and mesoporous (2–50 nm) range. This makes them ideal for their application as EDLC electrodes. Their ease of synthesis lowers the price to fabricate them. However, they suffer from some drawbacks. They are produced as flakes and as such are not freestanding. This requires the use of a polymer binder which further reduces the conductivity of the material. It is also overwhelmed by significant amounts of amorphous carbon that does not contribute effectively to the electrical conductivity of the material. Furthermore, this carbon-polymer mixture must be cast onto metal current collectors [27–30] to form electrodes, since in their free standing form they are not structurally stable. This makes them difficult to be used as wearable devices.

Carbon nanotubes (*CNTs*) (**Figure 2b**), have emerged as a very promising electrode material candidate because of their high theoretical property values. The individual multiwalled CNT (MWCNT) has been measured to have an average tensile strength of 60 GPa [31] and a Young's modulus of over 1 TPa [32, 33], with a low electrical resistivity of 3×10^{-5} ohm cm [34–36], and a thermal conductivity of 3500 Wm K^{-1} [37, 38]. All these properties arise from the sp^2 bonds connecting the carbon atoms in the form of a tube. However, as dispersed tubes, they would pose the same problems that traditional powdered carbon materials reveal. Hence, there has been extensive research on exploring ways to scale them up into macro-assemblages. This is directly related to their synthesis process. There are presently three
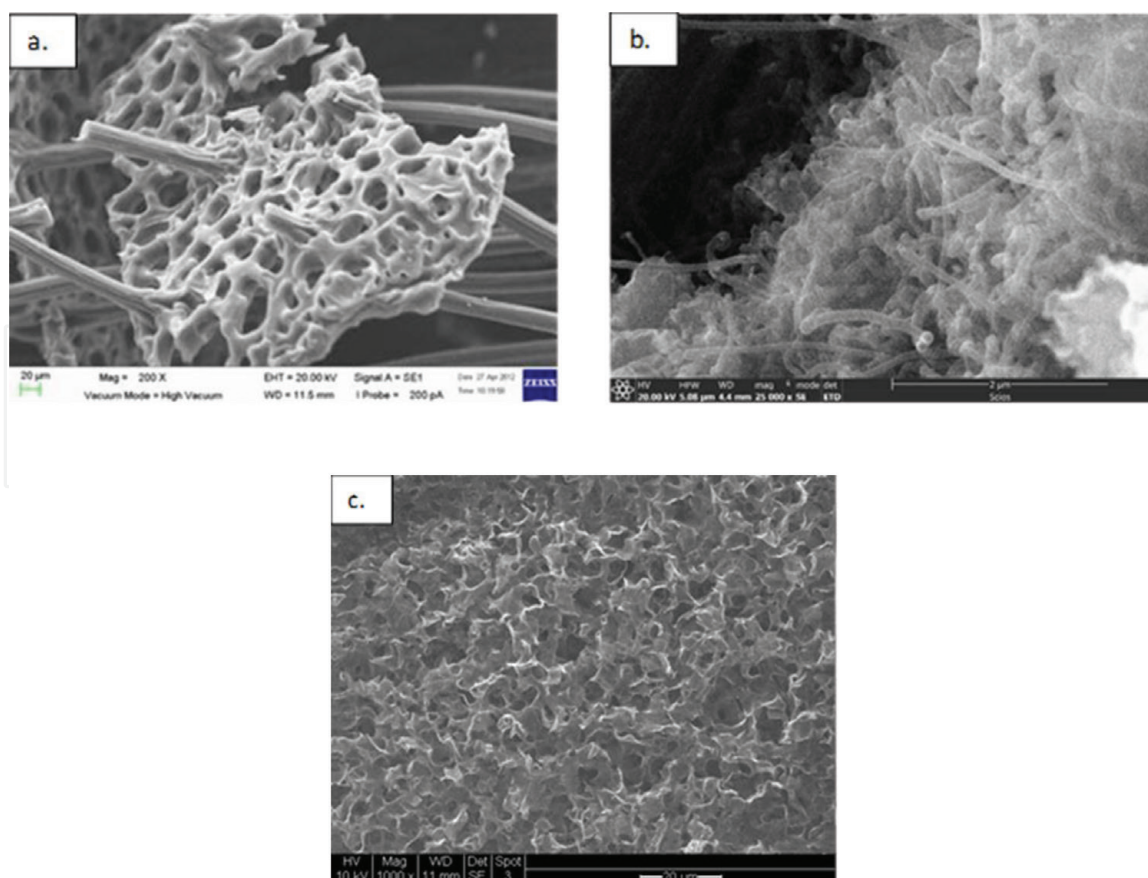
Figure 2.
SEM images of: (a) activated carbon, reprinted with permission from [45]; (b) CNTs and (c) graphene.

major established ones, namely direct assembly from gaseous phase, wet spinning and dry spinning from vertically aligned arrays. Macro-assemblages gathered from wet phase are drawn from lyotropic liquid-crystalline phase CNT matrix-which requires the use of corrosive acids to make the CNT solution [39–41]. This results in a final product that has already undergone post processing and contains catalyst impurities, which makes them less effective to further post processing treatments. The macro-assemblages drawn from vertically aligned (VA) CNT arrays, as made in a chemical vapor deposition (CVD) reactor, are in a much more pristine state. This makes them more ideal for further processing. The individual tubes are held together via van der Waals forces and hence require no binding material. This gives CNT assemblages the ability to act as free-standing structures and makes them a good candidate for wearable electrode material.

Graphene, **Figure 2c**, *and reduced graphene oxide (rGO) are* both highly researched allotropes of carbon for energy storage electrode material. However, as rGO is produced in the form of flakes, it requires a binder and thus cannot be very easily translated into a wearable device. Graphene provides the opportunity to be synthesized in a paper form. This, added with its high surface area and conductivity [11], makes it ideal candidate for a structurally stable current collector. CVD is the more versatile synthesis method as this allows for inclusion of various precursors for functionalization of the graphene. The catalyst can also be modified to tune the porosity of the graphene material. All of these tunable properties allow graphene paper to be used as a conductive housing for different active materials and electrolyte [42–44].

It is seen that sp^2 bonded carbon nanomaterials such as CNTs and graphene have the best suitable properties—high surface area, conductivity and strength—to elevate energy storage in the wearable format. Thus, they lend themselves as a pivotal milestone in the development of fabric-based energy storage devices.

Prior to establishing the norm of using carbon nanomaterials as conductive pathways/current collectors, *active materials* themselves have been used as electrodes. We define active material as those oxides, compounds or polymer chains that can store charge in the form of chemical reactions or volume expansions. However, due to their insulating nature and poor structural strength, their pristine form is mostly avoided for composite materials.

3. Dip coating as a tool for fabricating of fabric-based energy storage devices

Textile fabric materials made by weaving synthetic or natural fibers are highly flexible and possess high surface area. This makes them ideal templates for housing active materials that can store energy. Dip coating takes advantage of this characteristic by applying CNTs or graphene on the surface of the textile fibers via dipping in an aqueous solution that contains a dispersion of the active material (**Figure 3a** and **b**). Drying the fabric results in a coated material that is ready to be used as a flexible electrode. Cotton, owing to its strength and hydrophilicity, is primarily used for this process as it involves an aqueous solution. Hu et al., utilized this process to make CNT coated cotton fabric electrodes [46] (**Figure 3a** and **b**). They achieved high active loading of 8 mg/cm^2 resulting in a capacitance of 120 F/g. The impregnation and consequent coating is highly dependent on the permeability/retention ability of the textile used in this process. This renders dip coating a non-versatile and non-reproducible process. The density of packing is also affected making the material not highly conductive. To overcome these issues, screen printing was introduced. By the simple employment of a screen resist on top of the fabric before the application of a thick slurry of the active material, a binder-active material solution can be very evenly coated to the surface of the fabric textiles [46]. This allows for good control over exactly how much of active material is loaded depending on the concentration and volume of the slurry used. The method allows for multiple repetitions as an easy way to increase the mass loading. This way, a reproducible capacitance of 90 F/g was achieved [46].

However, there is an upper limit to the amount of active material that can be added because of the shrinking porosity of the fabric skeleton with impregnation. As a consequence, there is a maximum for the capacitance values that can be achieved. One factor that can be easily changed to better the properties is the active material. Pseudocapacitive materials can be used as active material known for their higher theoretical specific capacitance (F/g). Thus, with the same mass loading, much higher capacitances can be achieved. Yu et al. explored electrodeposition to coat manganese oxide onto graphene coated textile fibers [47], as shown in **Figure 4a**, which successfully increased the capacitance to 350 F/g. The same approach can be adopted to deposit other pseudocapacitive material, such as ruthenium oxide or
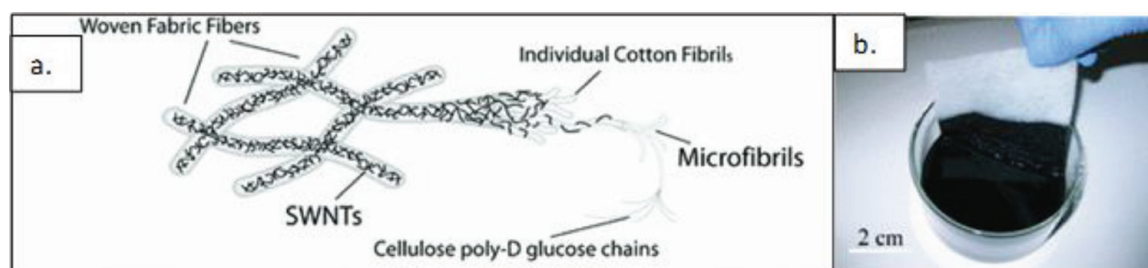
Figure 3.
(a) Schematic of screen printed CNT-cotton fibers and (b) picture of the dip coating process with cotton fabric. Reprinted with permission from [46]. Copyright 2010 American Chemical Society.
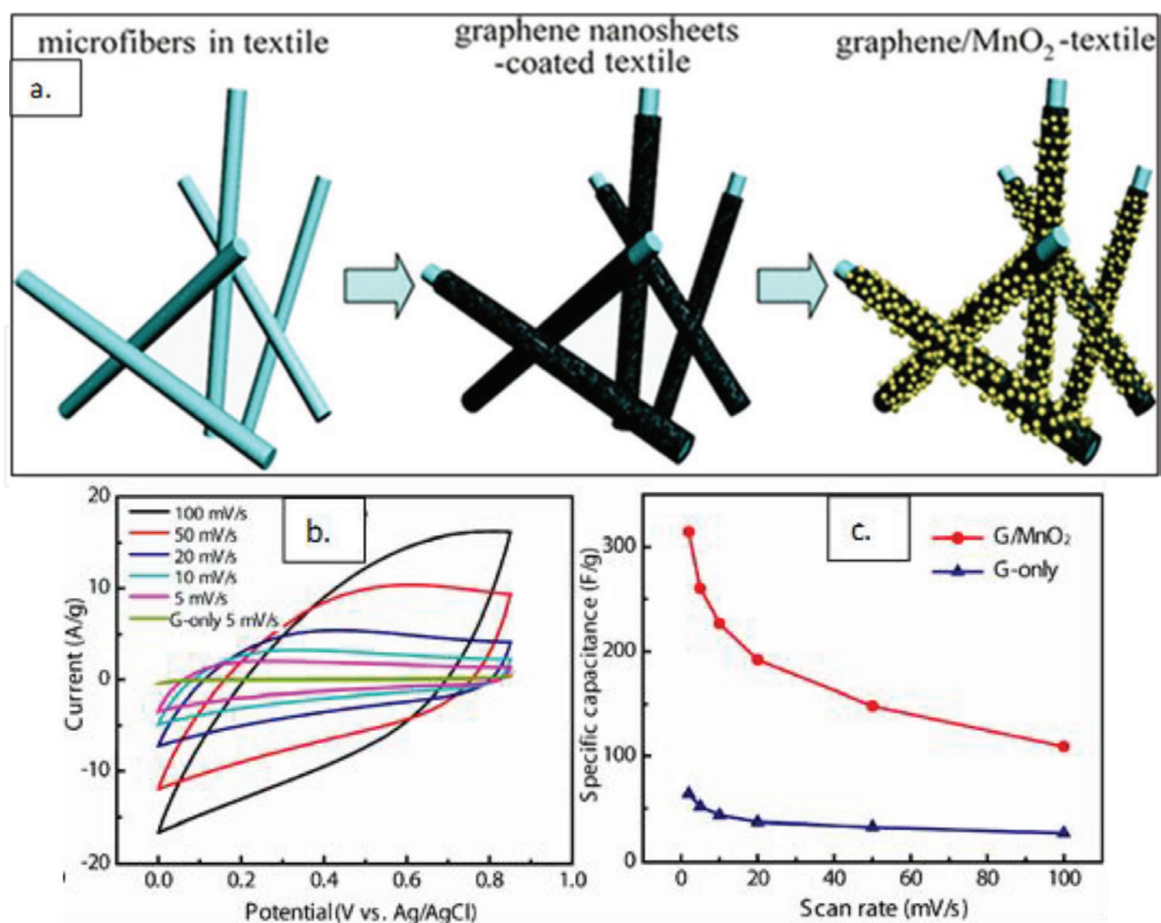
Figure 4.
(a) Schematic of electrodeposition of manganese oxide on graphene coated textile fibers; (b) half-cell cyclic voltammetry of manganese oxide—graphene—fabric electrode; (c) capacitance vs. scan rate curves. Reprinted with permission from [47]. Copyright 2011 American Chemical Society.

conductive polymers. An asymmetrical device with high energy densities can be fabricated by carefully choosing the electrodes based on their working reduction potential ranges. Changing the crystallinity and nanostructure of the deposited material by controlling the deposition parameters and composition can also yield better properties. The alpha phase of the manganese oxide is more conductive and electrochemically stable than its beta and amorphous forms. Thus, a very fine nanorod-structured deposition of alpha phase manganese oxide can greatly increase the energy density. Another option for the device property improvement is exploring more electrochemically active materials such as lithium metal oxides, sulfur, etc. Thus, with the use of a polymer electrolyte matrix separator, a flexible battery device can be fabricated.

As demonstrated in the cyclic voltammetry curves in **Figure 4b** and **c**, there is still a very non-capacitive (rectangular) nature to the fabric devices. This is in part a contribution from the low loading of conductive material—CNT or graphene. The other active materials (pseudocapacitive or battery) are not very conductive in nature and highly rely upon the carbon-base material for their electron transfer requirements. Thus, there are heavy losses in terms of power density in such materials. This can be avoided with the use of metals as current collectors, however making the device non-flexible.

The most promising advantage of the discussed dip coating is the low production cost and time consumption. However, further improvement of the carbon coating material needs to be realized to progress in this fabrication method. An alternative is a fully core to shell design made completely of conductive carbon material and coated active material. This would result in a 100% mass loading of active material and would give more energy per unit gram of fabric.

4. Fiber-based electrodes

Electrodes in the form of yarns that can be readily stitched as a textile fabric has the potential to overcome many of the disadvantages discussed in the previous section. The research in fiber/yarn-based electrodes began with dip coating techniques and as discussed in the sections before, some challenges have emerged. The conductivity and active loading of the material was low, and the related research eventually moved towards more promising approaches. One of them is the possibility to realize a fully active material loaded fiber form.

A conductive carbon-based core that provides the strength, conductivity and flexibility with an outer shell of active material that secures high energy density has been explored. CNTs dispersed in acid and extruded in the form of a fibers which have been easily fabricated. These porous fibers act as a current collector and housing for the active material. Vertically aligned CNT arrays can be used to spin long and porous fibers. These are produced in a more pristine format and hence can be easily processed and react less with electrolytes. These full core-shell carbon structures have very high surface area ~1400 m^2/g, which gives the opportunity to take advantage of the ELDC characteristic of these fibers [48]. Novel fiber structures utilizing higher surface area graphene have also been fabricate. RGO based fibers that are highly porous and boasting surface area up to 2400 m^2/g have been reported, which results in a very high energy density of 400 F/g [49] (**Figure 5i a–c**). Hybrids involving both CNTs and graphene have been synthesized, utilizing the high surface area of both material to create a sponge like fiber material [48] (**Figure 5ii a–c**). Flexible fibers were successfully woven into a fabric and its performance remained consistent through exposure to bending stress. The major drawback of these high energy storage materials is their strength. Although their Young's modulus is appreciable, which allows them to be flexible, their tensile strain to failure is quite low. This makes them prone to breakage and unfit to survive as a self-supporting fabric patch. Addition of any active material to the fiber would make them too brittle to handle.

Carbon fibers, which are a very sturdy and high strength materials, haves been used as a substrate to house pseudocapacitive manganese oxide [50] (**Figure 5v a-c**). The resulting fiber proved to be quite strong and showed appreciable energy storage values. However, the electrical conductivity of the material is still a concern. Conventional carbon nanotube fibers have also been used as conductive substrate for housing manganese oxide [51] (**Figure 5iv a–c**). The resulting composite was wound like a spring and encapsulated in a gel polymer, showing no losses in energy storage properties for tensile strains of up to 100%.

A polymer core with carbon nanomaterials grown/deposited across the surface derives most of its strength from the elastic polymer and can sustain higher tensile strains. Active material can then be successfully decorated on the surface [52] (**Figure 5v a–c**). This however suffers from the loss of conductivity and requires an additional current collector in the form of a multiplied metal yarn structure.

Fibers made completely out of electrochemically active material such as poly(3,4-ethylenedioxythiophene) (PEDOT) have also been developed [53]. The biscrolled PEDOT with the CNT matrix makes the composite quite easy to scale up. Further, due to the high electrical contact between the current collector and active material, the device shows good performance at 1 mV/s scan rate. The greatly twisted fiber boasts very high active material loading and high volumetric capacitance.

There are some challenges to the development of these forms of devices. They have the potential to be composed completely of active material leading to unsurpassed energy storage capabilities, however, the loss in strength of the material needs to be countered. A fiber that possess both great tensile strain, strength and high loading of active material could pave way to the next generation of wearable devices. We believe that the key lies in highly densified CNT fibers with modified surfaces.
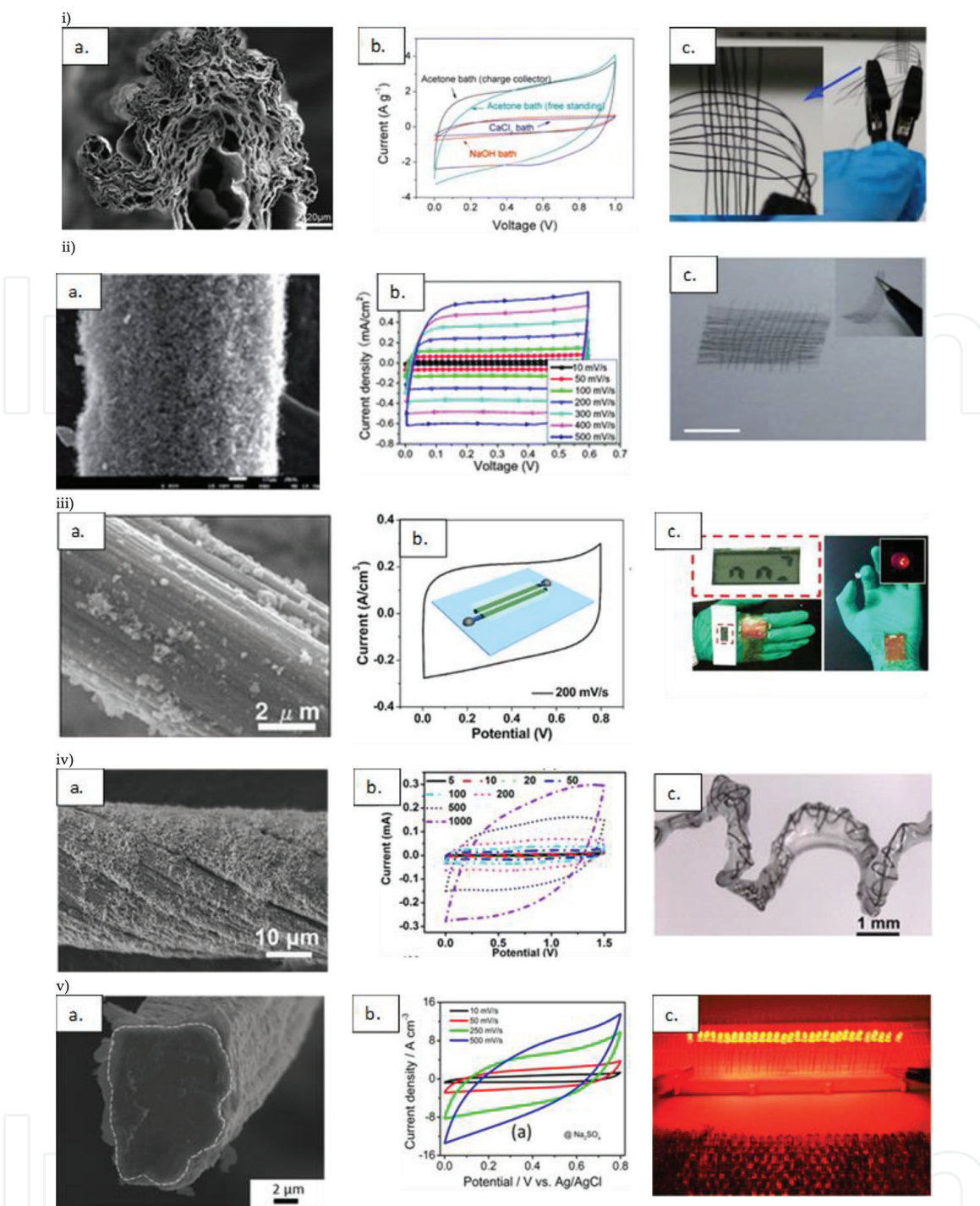
Figure 5.
(a) SEM image of electrode base material; (b) cyclic voltammetry graph of the working device; (c) picture of full device. (i) RGO fiber reprinted with permission from [49]. Copyright 2014 American Chemical Society. (ii) CNT-graphene fiber reproduced from [48] with permission from Royal Society of Chemistry. (iii) Carbon fiber coated with manganese oxide reprinted with permission from [50]. Copyright 2012 American Chemical Society. (iv) CNT fiber coated with manganese oxide reprinted with permission from [51]. Copyright 2015 American Chemical Society. (v) GO-active material coated stainless steel fiber reprinted with permission from [52]. Copyright 2015 American Chemical Society.

5. Ionic liquid-based fabric capacitor

Below are presented experimental results on ionic liquid-based fabric capacitors obtained by our team. A factor that is often ignored when it comes to wearable devices is the evaluation of their properties as a function of the weight of the materials. In these cases, most used is the area of the material or only the weight of the active material is taken into consideration. This becomes a limitation when scaling to industry production as bulky materials will not be suitable for powering

fabric-based devices. Most of literature data still relies on textile fibers to provide the structural and flexible skeleton for their electrodes. This requires the usage of two layers of fabrics (electrodes) that adds unnecessary weight to the full device which is mostly never reported. An aspect to consider is the synthesis approach. The more structured the active synthesized material, the higher the surface area and therefore gives a higher specific capacitance. A drawback of this is the cost—both monetary and time consuming—that scales exponentially with the structure complexity. The other major consideration that is overlooked is the practical feasibility of the devices fulfilling certain applications. Most wearable energy storage devices have adopted gel-based polymer electrolyte systems to create a flexible separator. However, they still rely upon aqueous electrolytes such as phosphoric acid, potassium hydroxide, etc. This caps the voltage window for conventional devices with high cyclability to a low value of 0.8 V. However, even LEDs require around 1.3 V to operate. The non-conventional asymmetrical devices can push the envelope to around 1.6 V, but beyond that they suffer from heavy capacitance losses. In this section, we hope to provide a solution based on our work to these practical challenges.

Aqueous electrolytes, although inexpensive and stable, are limited by their low reduction potentials with most conventional carbon materials. Anything beyond voltage window of 1 V results in irrecoverable losses in both the electrode material and the electrolyte. This limitation comes mainly from the presence of water. Thus, a change from aqueous to organic solutions results in a drastic change in the voltage window stability range. Commonly explored solutions are ethanol with suitable salts such as lithium perchlorate dissolved in them. Devices made with this electrolyte can expand the operation window from 1 V to around 2.2 V. This window decreases with introduction of pseudocapacitive materials to around 1.2 V, which is much higher than the 0.8 V offered by aqueous electrolytes. A device with asymmetrical electrodes can increase this window up to 2.2 V. To achieve a higher voltage window more expensive and less explored materials need to be considered. The latter are known as ionic liquids (ILs). These are a highly reactive group of electrolytes that are a combination of an organic cation with a very electronegative anion. Some of them—1-ethyl-3-methylimidazolium tetrafluoroborate ($EMIMBF_4$)—have a very large and stable working voltage window, going as high as 4.5 V. They find usage as catalysts for higher voltage reduction reactions, however there is an emerging interest in their utilization in energy storage applications. ILs are most commonly combined with polyvinylidene fluoride-hexafluoropropylene (PVDF-HFP) to create a gel polymer electrolyte separator matrix.

We introduce here a novel way to incorporate the IL $EMIMBF_4$ into any fabric material (from stretchable polyester sports-wear to sturdy military fabric) that utilizes only the essential amount of fabric material. For our tests we procured normal Bucky paper (BP) from General Nano, LLC which is a matrix of single walled carbon nanotubes under no alignment. Our procedure includes using the fabric as a separator to house the gel electrolyte and eventually be sandwiched between symmetrical BP electrodes (**Figure 6a**). Initially, polyvinyl alcohol (PVA)-sulfuric acid gel matrix was used. The gel electrolyte (1 g of PVA in 10 ml 0.1 M sulfuric acid) was dropped into the fabric. The soaked fabric was then allowed to dry till it reached a gel-like consistency. The electrodes were then applied on top of the electrolyte and allowed to further dry until they became integrated into the fabric electrolyte structure. This device was tested electrochemically using a Potentiostat Interface 1000, Gamry Instruments and showed results comparable to conventional devices made without the fabric separator. A similar gel polymer concentration was utilized for the IL (3 ml $EMIMBF_4$ in 10 ml acetone with 1 g PVDF). The same methodology was adopted to create the gel fabric. However, the electrochemical results with the device were not comparable with average values of conventional devices. This was
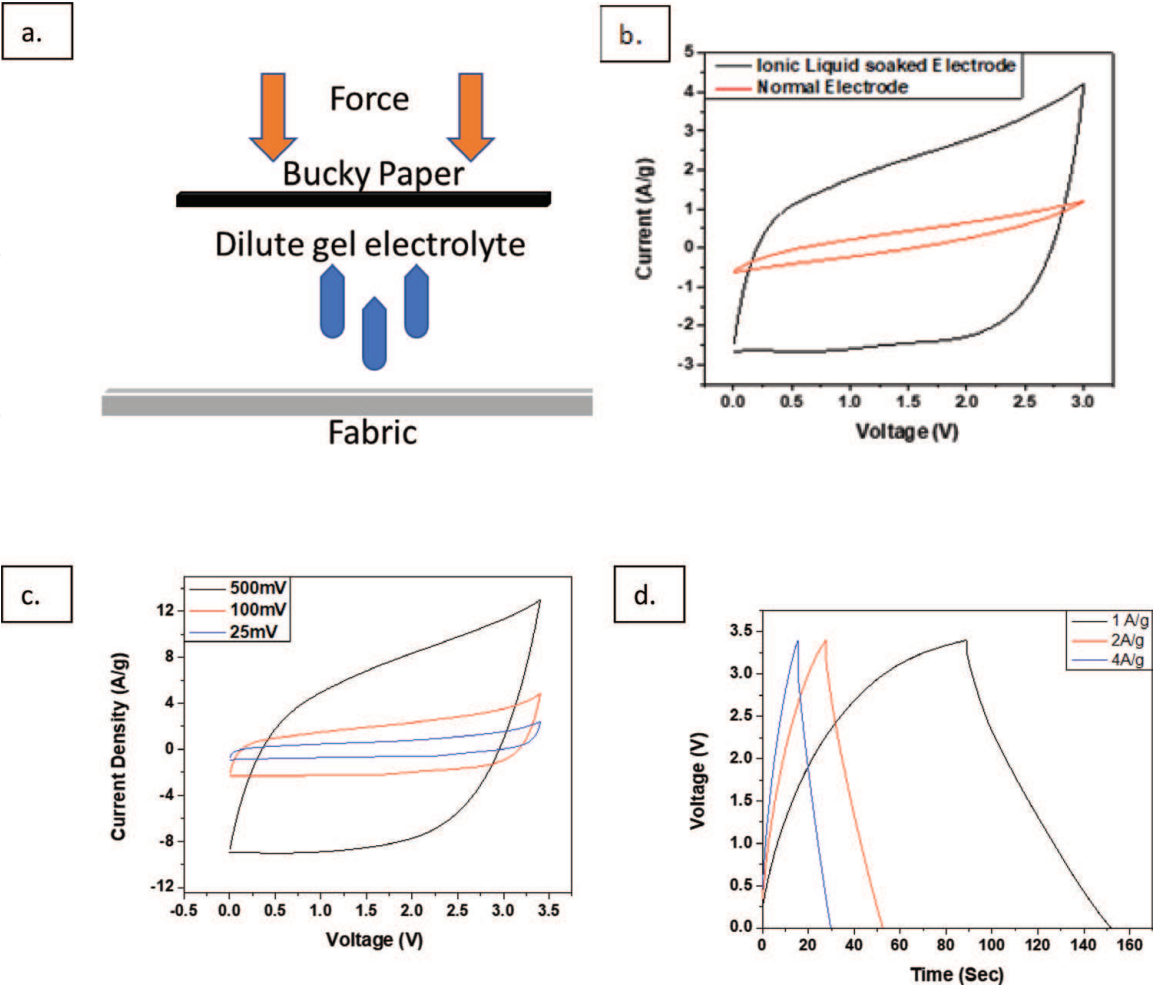
Figure 6.
(a) Schematic of creating gel fabric structure with Bucky paper electrode pressed on top; (b) cyclic voltammetry curves at 200 mV/s for normal BP electrode and BP electrode pre-soaked in IL solution; (c) different scan rate cyclic voltammetry curves of IL-soaked device; and (d) charge and discharge curves of the fabricated device obtained at different current densities.

not an indication of the limit of the electrode as the sulfuric acid device gave appreciable values. We added a further step to the device fabrication, which included first soaking the electrode in a solution of the IL plus acetone, and then incorporating it into the fabric structure. This resulted in an increase of the device energy density (**Table 1** and **Figure 6b–d**). Since acetone is fast evaporating solvent, it did not allow for the electrode to be fully penetrated with the electrolyte before drying. The described soaking procedure ensured complete utilization of the electrode surface area and thus increased the stored energy.

The device can be steadily cycled between 0 and 3 V window range without fade in capacitance values. This is not the only major advantage of such a device. Since

Gel electrolyte	Specific capacitance (F/g)	Energy density (Wh/kg)
PVA–sulfuric acid (1 V)	11.54	3.2
PVDF–ionic liquid (3 V)	61.66	19.53
Ionic liquid soak	137.6	48.62

Table 1.
Specific capacitance and energy density values of the fabric device achieved with different polymer electrolytes and preparation techniques.

the fabric is being used as a separator and not as the electrode itself, it essentially reduces the weight of the device in half, as compared to other wearable energy storage devices that rely on a fabric skeleton backing for each electrode. The fabric can then be further sown in as a patch thus making the only additional weight contribution to the device that of the gel electrolyte. Another key advantage of the described system is the ability to be easily modified for various applications. Depending on the energy need, the area of the electrode used can be increased or decreased to give highly tunable capacitance values (**Figure 7a** and **b**). Since the application and fabrication of the gel fabric is easily controlled, the device can also be made into sections. Various electrodes can be applied next to each other as isolated devices on the same fabric. One patch of fabric could contain many isolated devices. This gives high tunability to the device. Three isolated devices were made in one patch and when connected in series it achieved a working voltage of 9 V. In a parallel wiring
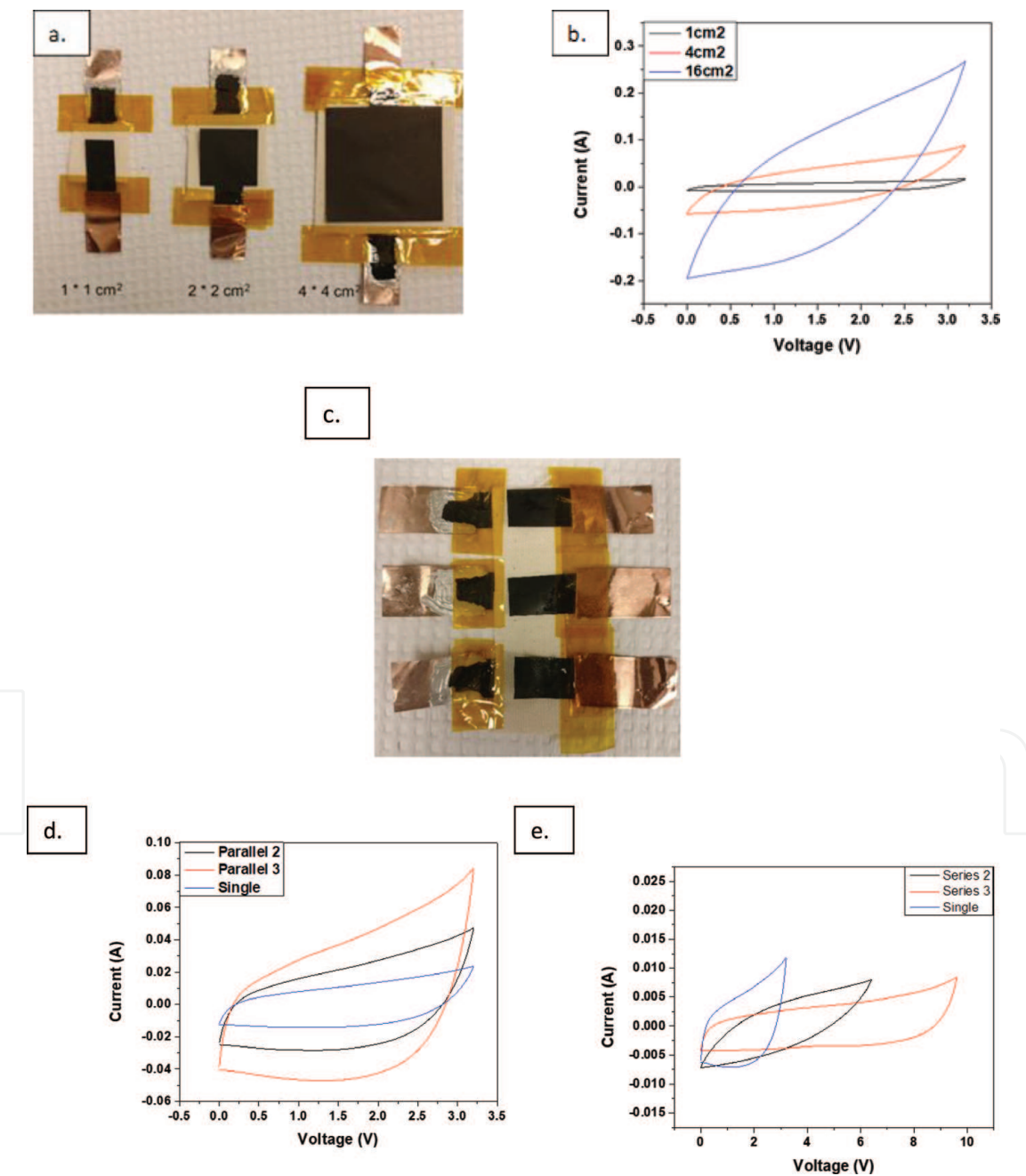
Figure 7.
(a) Pictures of different size with increasing areas of electrode materials used for the fabric device; (b) corresponding cyclic voltammetry profiles at 200 mV/s scan rate; (c) picture of isolated devices in a single patch of fabric; (d) corresponding cyclic voltammetry profiles at 200 mV/s scan rate in parallel connections and (e) corresponding cyclic voltammetry profiles at 200 mV/s scan rate in series connections.

the device achieved 3 times the current output and energy density of an individual one (**Figure 7c–e**). The fabricated device is highly versatile in terms of how it can fit to different applications. Electrical contacts can be made by simply applying copper tapes with silver paint to the ends of the electrodes. These connections need not be pre-made, giving a further degree of freedom.

Textile fabrics are exposed to stress in the form of flexing, bending or wrinkling. This is unavoidable in clothing when the wearer executes any form of motion. Such an environment must be taken into consideration when making fabric integrated devices. Here we tested our fabric device by exposing them to 10,000 cycles, where in each cycle it was bent to an angle of 120° and brought back to normal shape (**Figure 8a**). This is a higher degree of bending than fabrics
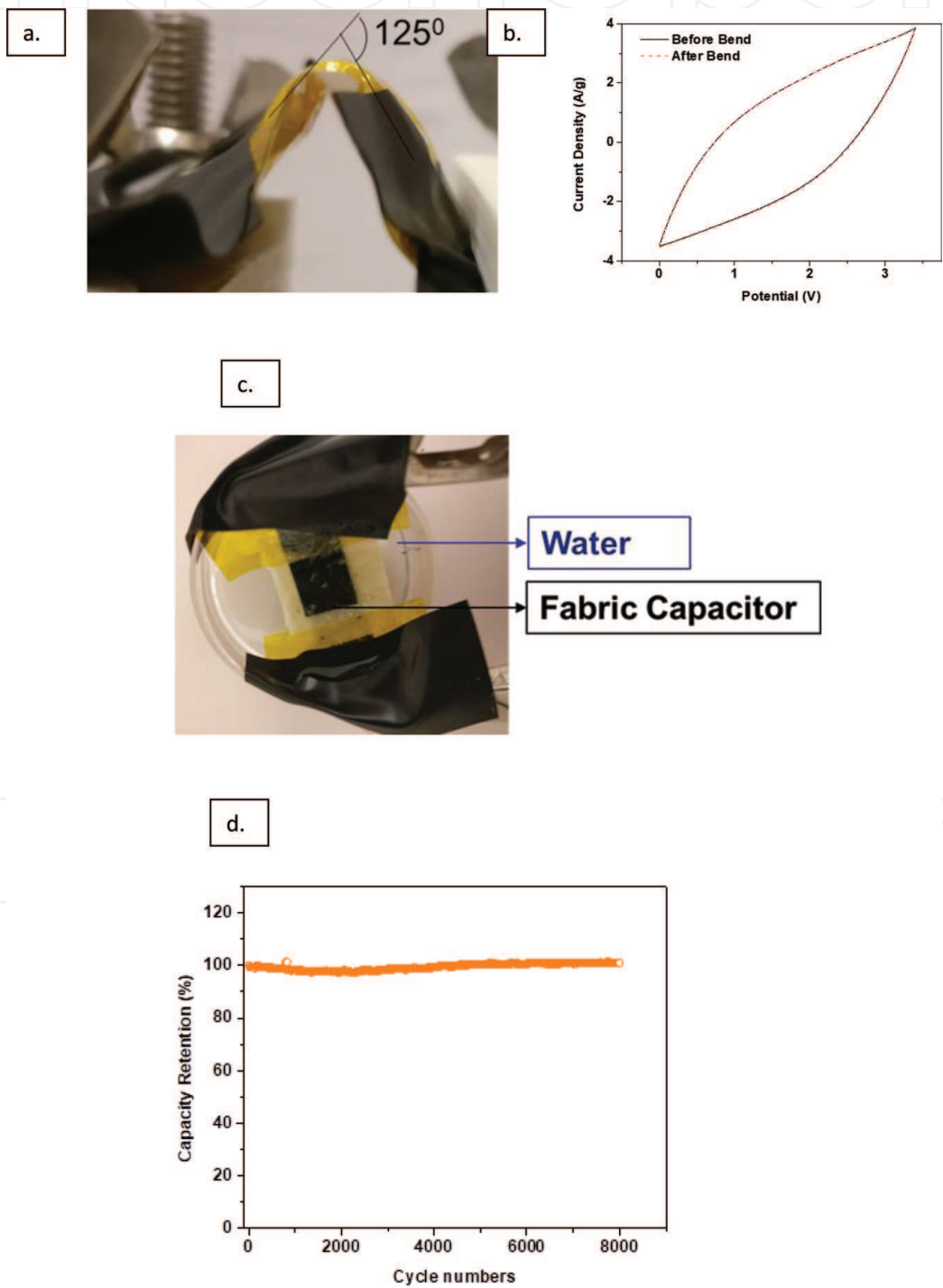
Figure 8.
(a) Picture of the degree of bending during each cycle of the stress test; (b) cyclic voltammetry curve of the device before and after bending; (c) picture of the water bath immersion tests; and (d) cycling curve of the device at 2 A/g current density.

would have to usually sustain on a daily basis. The device showed no depreciation in terms of capacitance after the cycling (**Figure 8b**). Another consideration to be taken into account is the exposure to moisture in the form of sweat from the wearer. Our device was soaked in a bath of water and cycled. In this test no observable depreciation in energy density was noticed (**Figure 8c**). Finally, the device was cycled 8000 times at a current density of 2 A/g that resulted in a final capacitance of 102% (**Figure 8d**). This increased capacitance is due to the forcible opening of the pores of the BP electrode with constant cycling. The high retention rate is very promising and ensures that the electrolyte is not being degraded at such high voltage ranges.

There still exists room for improvement for the described architecture of the fabric devices. Altering the electrodes by creating more energy dense materials is a quick way to further improve the properties of the device. Using pseudocapacitive materials like transition metal oxides or conductive polymers in an asymmetrical electrode arrangement has the potential to further enhance the properties. This will be explored in our following works.

6. Conclusion

The wearable energy storage devices have seen much development over the last decade. This evolution included starting from cotton coated electrodes that were not highly reproducible and moving to more reliable techniques such as screen printing using a polymer resist. Further, full active material loaded yarns have been realized. There are however drawbacks with each of the approaches which include either a limit to the active material content and subsequently the energy density, or a lack of strong materials that can sustain the wear and tear of fabrics that will potentially go into articles of clothing. The most promise is shown when using yarn-based devices that, if coupled with high strength, may provide the best properties.

In our work we designed an approach to making a tunable device that can accommodate a various range of practical applications including those requiring voltages as high as 9 V or high current ratings. The gel fabric separator fabricated and reported here utilizes just the essential amounts of fabric, which can be easily patched onto articles of clothing thus adding no extra weight. The final device we created exhibits an energy density of 48 Wh/kg at 1 A/g current density and can sustain high cycling without any noticeable losses in capacitance. The described fabrication approach is versatile and can be adopted for making various electrodes that can further improve the properties of the fabric energy storage devices.

Acknowledgements

Conflict of interest

The authors declare that there was no conflict of interest during this work.

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