

Reduced graphene oxide and polypyrrole/reduced graphene oxide composite coated stretchable fabric electrodes for supercapacitor application



Chen Zhao, Kewei Shu, Caiyun Wang^{**}, Sanjeev Gambhir, Gordon G. Wallace^{*}

Intelligent Polymer Research Institute, ARC Centre of Excellence for Electromaterials Science, University of Wollongong, NSW, 2522, Australia

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ABSTRACT

The advent of self-powered functional garments has given rise to a demand for stretchable energy storage devices that are amendable to integration into textile structures. The electromaterials (anode, cathode and separator) are expected to sustain a deformation of 3% to 55% associated with body movement. Here, we report a stretchable fabric supercapacitor electrode using commonly available nylon lycra fabric as the substrate and graphene oxide (GO) as a dyestuff. It was prepared *via* a facile dyeing approach followed by a mild chemical reduction. This reduced graphene oxide (rGO) coated fabric electrode retains conductivity at an applied strain of up to 200%. It delivers a specific capacitance of 12.3 F g^{-1} at a scan rate of 5 mV s^{-1} in 1.0 M lithium sulfate aqueous solution. The capacitance is significantly increased to 114 F g^{-1} with the addition of a chemically synthesized polypyrrole (PPy) coating. This PPy-rGO-fabric electrode demonstrates an improved cycling stability and a higher capacitance at 50% strain when compared to the performance observed with no strain.

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1. Introduction

Smart textiles, or E-textiles, are fabrics with embedded functional devices such as sensors, transistors or antennas [1–4]. This has gained tremendous interest in recent years and found widespread application in personalized electronics, wearable medical monitors, and even in the military field [5]. Textile based energy storage devices are needed to power such devices, since conventional batteries and supercapacitors are too bulky, rigid and heavy to be seamlessly integrated into wearable garments [6]. Compared to batteries, supercapacitors possess advantages in high power density, fast charging-discharging and extremely long cycling life [7]. Self-powered garments can be created by combining them with textile piezoelectrics [8] or thermoelectrics [9] to capture energy from body movement or heat respectively.

These structures are expected to accommodate an average deformation of 3% to 55% due to body movement [10]. There exists three main strategies to develop textile supercapacitors [11]: using

coated textiles as electrodes [6,12–20], fibres or yarns integrated into non-woven [21] or woven [22] textiles as electrodes, and fibre supercapacitors woven into textiles [23–28]. Among these approaches, coating the pre-existing textile structures with active electrode materials is the most facile and cost-effective technique [6].

Recently, diverse carbonaceous materials, such as single-walled carbon nanotubes (SWNTs) [12,14], activated carbon [6,16] and graphene [13,15,20] have proven to be useful in fabricating coated textile electrodes. Conducting polymers, such as polypyrrole (PPy) [17–19], have also been used to produce textile based electrodes. A stretchable supercapacitor assembled with activated carbon coated textile electrodes exhibited a $\sim 30\%$ capacitance loss with 50% strain applied [16]. SWNTs coated textile electrodes could sustain a 120% strain without obvious capacitance fade [12], and the chemically or electrochemically synthesized PPy/fabric electrodes even showed improved performance while being stretched [17,18]. For large-scale applications, cost reduction of SWNTs is still required [13], and poor cycling stability of PPy should also be improved.

Graphene is a promising alternative to construct textile electrodes, as it can be produced from inexpensive graphite at a large-scale via cost-effective chemical methods. Recently, graphene based textile electrodes demonstrated high capacitance and flexibility [13,15,20]. Solution processed graphene coated textiles

^{*} Corresponding author. Tel.: +61 2 42213127; Fax: +61 2 42213124.

^{**} Corresponding author. Tel.: +61 2 42981426; Fax: +61 2 42983114.

E-mail addresses: caiyun@uow.edu.au (C. Wang), gwallace@uow.edu.au (G.G. Wallace).

delivered a specific capacitance of $\sim 63 \text{ F g}^{-1}$ [13]. A high specific capacitance of 326.8 F g^{-1} was achieved by a thermally annealed graphene coated cotton cloth [15]. Graphene coated electrospun polyamide-66 nanofabrics exhibited a specific capacitance of 280 F g^{-1} [20]. However, the stretchability of those textile electrodes was not discussed.

In this work, a stretchable conductive fabric electrode was prepared using GO as a dyestuff and nylon lycra fabric as the substrate. A facile dyeing approach followed by a mild chemical reduction was employed. An additional coating of redox-active and highly conductive PPy film will further enhance the performance of this fabric electrode. PPy can be integrated with graphene nanosheets through the π - π stacking interaction between the rGO sheet and the conjugated backbones of PPy [29,30]. Currently reported PPy/rGO composites are either in powder or sheet form, they can't be stretched. Here, PPy was chemically polymerized onto the conductive rGO coated fabric to achieve a stretchable PPy/rGO/fabric composite electrode. rGO sheets play the role of stabilizer for PPy, improving the poor cycling stability of PPy as described in our previous work [17]. The electrochemical properties of these fabric electrodes under strain were investigated.

2. Experimental

2.1. Reagents and materials

Nylon lycra fabric was purchased from Spotlight Stores Pty Ltd. Pyrrole (Py) was sourced from Merck. Naphthalene-2,6-disulfonic acid disodium salt (Na_2NDS), ammonium persulfate (APS) and lithium sulfate monohydrate ($\text{Li}_2\text{SO}_4 \cdot \text{H}_2\text{O}$) were obtained from Sigma-Aldrich. L-ascorbic acid (L-AA) was purchased from Alfa-Aesar. Pyrrole was freshly distilled, whereas other chemicals were used as supplied. All aqueous solutions were prepared using Milli-Q water ($\sim 18 \text{ M}\Omega$).

2.2. Preparation of rGO coated fabric

Graphite oxide was synthesized from graphite flakes using the modified Hummers' method [31,32]. Graphite oxide was exfoliated to graphene oxide using a Branson Digital Sonifier (S450D, 30% amplitude, 2 s on and 1 s off) for 30 min, forming a stable suspension with a concentration of 3 mg mL^{-1} . The nylon lycra fabric was treated with an aqueous solution of scouring agent and 10% sodium hydroxide for 1 h, followed by rinsing with abundant water. After drying, it was treated with plasma to increase the wettability prior to the dyeing process.

The fabric ($1 \times 3 \text{ cm}^2$) was immersed into a GO suspension for 30 min at room temperature and then dried at 60°C for 30 min. This process was repeated to increase the GO uptake. The reduction of GO coated fabric was performed by immersing into 60 mL of 0.1 M L-ascorbic acid solution (95°C) for 60 min with constant stirring. Such dyed fabric was washed with abundant water and finally dried at 60°C under vacuum overnight. The resultant fabric is denoted as rGO-fabric.

2.3. Preparation of PPy modified rGO-fabric

PPy was chemically polymerized onto the rGO-fabric according to our previous report with minor modification [17]. Briefly, rGO-fabric ($1 \times 3 \text{ cm}^2$) was immersed in a 20 mL aqueous solution containing Py and Na_2NDS . 5 mL ethanol was added to improve the wettability of rGO-fabric. The solution was kept stirring in an ice bath for 30 min, followed by addition of 5 mL pre-cooled APS

solution drop-wise to initiate polymerization. The reaction was performed in an ice-bath for 0.5 to 3 h. The concentration of Py, APS and Na_2NDS was 0.02 M, 0.02 M and 0.009 M, respectively. The coated fabric was washed with abundant water, followed by sonication in water for 30 min to remove the loosely bound PPy. Finally, the fabric was dried at 40°C under vacuum overnight. The obtained fabric composite is coded as PPy-rGO-fabric. The fabric electrodes with 2 h PPy deposition showed the highest specific capacitance. Only these samples were chosen for the investigation in this work. The loading amount of PPy was $\sim 2.8 \text{ mg cm}^{-2}$.

2.4. Characterization

Physicochemical properties: XRD patterns were recorded by a powder X-ray diffraction system (GBC MMA, Cu $K\alpha$ radiation, $\lambda = 0.15418 \text{ nm}$). Raman spectra were obtained using a confocal Raman spectroscope (Jobin Yvon HR800, Horiba) utilizing 632.8 nm diode lasers. GO was drop-casted on the substrate (quartz for XRD and glass slide for Raman) and dried for the characterization. The morphology of the fabric electrodes was characterized by FE-SEM (JEOL JSM-7500FA). The surface resistivity of the fabric electrode was measured according to the American Association of Textile Chemists and Colourists Test Method 76-1995 [33]: two rectangular copper electrodes were placed on the sample and the surface resistivity (ρ_s) was given by:

$$\rho_s = R \times \frac{L}{D} \quad (1)$$

Where ρ_s is the surface resistivity ($\Omega \text{ sq}^{-1}$), R is the surface resistance (Ω) measured by the multimeter, L is the length of the copper electrode and D is the separation distance between the copper electrodes.

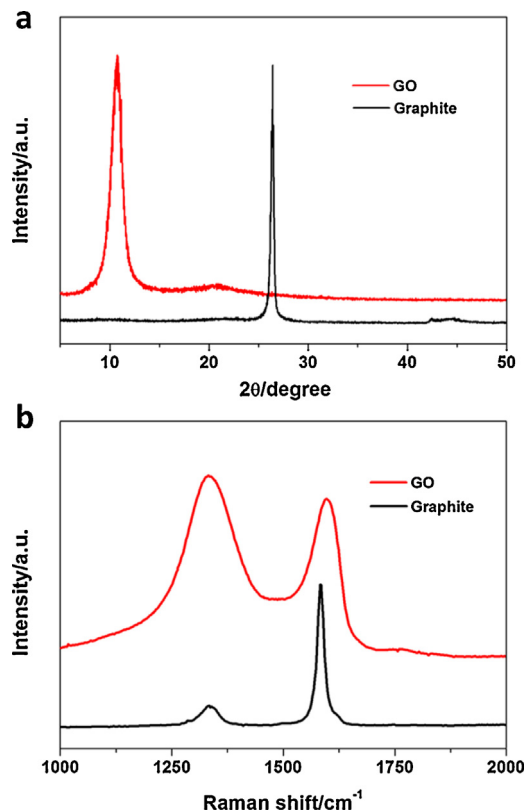


Fig. 1. XRD patterns (a) and Raman spectra (b) of GO and graphite.

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