Nonconductive commercial viscose yarns have been coated with a commercial conducting poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS) layer providing electrical conductivity which allowed a second coating of the electroactive conducting polymer polypyrrole through electropolymerization to develop textile yarn actuators. To simplify the PEDOT coating process and at the same time make this process more suitable for application in industry, a new coating method is developed and the properties of the PEDOT-PSS conducting layer is optimized, paying attention on its effect on the actuation performance. The effect of the concentration of an additive such as dimethylsulfoxide (DMSO) on actuation, and of PEDOT:PSS layers, is investigated. Results show that on improving this conducting layer, better performance than previously developed yarn-actuators is obtained, with strains up to 0.6%. This study provides a simple and efficient fabrication method toward soft, textile-based actuators for wearables and assistive devices with improved features.

1. Introduction

There is a need to develop new actuators that are soft, compliable and silent, and that provide natural motion to mechanically active wearables such as orthotic devices and exoskeletons. Different technologies have been proposed for this aim. Pneumatic actuators have been embedded into a sock-like device for ankle–foot rehabilitation. Another ankle–foot prosthetic active device has been developed using variable stiffness materials by combining thermally responsive polymers such as polyactic acid (PLA) and a conducting material such as graphene. Prosthetic assistive devices for other parts of the body have also been developed; for example, a glove for the hand powered by pneumatic actuators that can be used for assistance and rehabilitation, or an assistive orthotic device for the elbow and garments providing pressure to the upper part of the body, the latest two examples driven by shape memory alloys.

An interesting way to achieve soft actuators for wearables is by combining advanced textile processing, including knitting and weaving, with electroactive polymers that provide mechanical actuation. This type of technology is called textile actuators.

Conducting polymers (CPs) are electroactive polymers that deform under an electrical stimulation due to a reversible electrochemical reaction changing material composition and properties (such as volume). These polymers have been used as soft actuators and often called artificial muscles due to their functional similarity with natural muscles.

Of the CPs used in actuators, polypyrrole (PPy) doped with dodecylbenzenesulfonate (DBS⁻) is often used due to its good mechanical properties and the large volume change. The volume change is mainly caused by the insertion or ejection of cations and solvent into the polymer matrix (Figure 1) according to the following reaction (1):

$$\text{PPy}^0(\text{DBS}^-)_n(S)^+ (\text{S})_m \leftrightarrow \text{PPy}^{n+}(\text{DBS}^-)_n$$

$$+ n(C^+)_{aq} + m(S) + n(e^-)$$

where PPy⁰ represents the neutral polymer chains, DBS⁻ represents the macroanion trapped inside the CP during electropolymerization, C⁺ are cations exchanged during oxidation/reduction, m are the number of solvent molecules (S), n is the number of electrons, and e⁻ are the electrons.

When PPy is oxidized (becoming PPyⁿ⁺) under the flow of n electrons (e⁻), positive charges in the PPy chains (polarons and bipolarons) compensate negative charges from the trapped DBS⁻ anions and cations and solvent are expelled from the PPy into the solution to keep charge and osmotic balance and the material...
shrink. When PPy is reduced, cations and solvent molecules penetrate the material to compensate for the net charge of the trapped DBS– anions and PPy swells.\textsuperscript{[14]}

There are several factors influencing CP actuation. A wider voltage range applied to the polymer contributes to larger actuation strain, although too high values could promote the degradation of the CP by overoxidation or the presence of parallel reactions that may affect the oxidation/reduction of the CP originating the movement.\textsuperscript{[15,16]} Another main factor is the ion transport. The actuation strain rate is influenced by the speed at which ions move through the polymer.\textsuperscript{[17,18]} Moreover, it has been established that thin actuators exhibit much higher strain rates compared with thick actuators at the same potential applied as the diffusion path lengths are shorter, resulting in faster actuation.\textsuperscript{[19,20]} The type of solvent (e.g., water, acetonitrile, or propylene carbonate) is also a factor that determines the performance of the material.\textsuperscript{[21]} Thus, to achieve fast actuation, thin layers or thin fibers have to be used.\textsuperscript{[22]} Yarns coated with relatively thin PPy layers should result in fast actuation; however, they will deliver low forces. By combining CPs with textiles, every CP-coated yarn will act in a collaborative way to achieve an amplification of force and strain.\textsuperscript{[7]}

Poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS) is another well-studied CP due to its structural stability, processability, and high electrical conductivity along with optical transparency.\textsuperscript{[24]} Applications that have been proposed include printed electronics,\textsuperscript{[24]} charge storage devices,\textsuperscript{[25]} solar cells,\textsuperscript{[26]} and textile electronic devices such as wires to connect electronic elements\textsuperscript{[27]} or fabric-based thermoelectric modules.\textsuperscript{[28]} The use of polar organic solvents such as ethylene glycol (EG), methanol, and dimethylsulfoxide (DMSO) has been shown to increase the conductivity of PEDOT:PSS thin films.\textsuperscript{[29–31]} According to the literature, DMSO results in a better increase in the conductivity of PEDOT:PSS.\textsuperscript{[32]} However, the mechanism by which the organic solvent acts on the conductivity of PEDOT:PSS is still debated. Some authors have reported that the post-treatment of a PEDOT:PSS film by immersion in a polar organic solvent turns the film insoluble and improve the conductivity by removing PSS.\textsuperscript{[13,34]} Others mention that the structure of PEDOT chain changes from benzoid to quinoid structure after DMSO treatment.\textsuperscript{[29,33]} In others words, the DMSO may act as secondary dopant, which improves the morphology of PEDOT:PSS and increases the conductivity of the film, which depends on the concentration of DMSO.\textsuperscript{[35]}

In this study, we used commercially available PEDOT:PSS as a first coating to provide textile yarns with a conducting layer that will be used to perform a subsequent electropolymerization of actuating PPy to fabricate yarn actuators for textiles. The performance and deposition of such PEDOT:PSS conducting layer has been investigated regarding the number of coatings and the addition of DMSO at different concentrations (the most successful additive so far to improve PEDOT:PSS conductivity)\textsuperscript{[34]} to get high-conducting PEDOT:PSS and high-strain yarn actuators. Using PEDOT:PSS from solution instead of as previously vapor phase polymerized PEDOT, will simplify the coating procedure, which, in addition, is compatible to large-scale production methods used in textile manufacturing as it is similar to currently used dyeing methods.\textsuperscript{[36]}

2. Results

2.1. PEDOT:PSS Coating

The viscose yarns were coated with PEDOT:PSS as shown in Figure 2 and Section 4 (Methods) and thereafter checked by visual inspections, scanning electron microscopy (SEM), and energy-dispersive X-ray spectroscopy (EDX). Figure 3 shows the photo and SEM images before and after the PEDOT:PSS coating, as well as EDX spectroscopy of the PEDOT:PSS coated yarns.

In Figure 3a, the presence of PEDOT:PSS coating on the yarn can be clearly observed due to its dark blue color.\textsuperscript{[37]} The SEM images (Figure 3b,c) show no significant differences, which indicates a nice uniform, conformal coating of the PEDOT:PSS on the viscose yarns.

EDX spectroscopy was used to characterize the chemical composition of PEDOT:PSS coated yarn. Figure 3d shows major peaks of C at 0.27 keV and S at 2.307 keV characteristics elements.

![Figure 1. Scheme of electrochemical redox reaction of PPy doped with large, immobile anions (symbolized by orange spheres), causing change volume due to out and influx of cations (yellow spheres) and solvent molecules (blue spheres).](image-url)
in PEDOT:PSS. A smaller peak at 1.04 keV regarding Cu could be detected, as Cu tape was used to attach the yarn to the microscope support. When the EDX spectra were obtained from the bare uncoated yarns, no S peak could be observed, pointing to the fact that S originates from the successful PEDOT:PSS coating.

### 2.2. Effect of DMSO Concentration on the Conductivity of PEDOT:PSS Coated Yarns

Next, we investigated the effect of adding DMSO at various concentrations to the PEDOT:PSS dispersion to increase the conductivity. The viscose yarns were coated with PEDOT:PSS and DMSO ranging from 0 to 80% (v/v), as stated in the Methods section, and the conductivity was measured. Figure 4a shows the conductivity of PEDOT:PSS coated yarns as a function of DMSO concentration. It is observed that when no DMSO was used, the conductivity of the yarn was low (~0.5 S cm$^{-1}$). After adding DMSO, the conductivity increased by three orders of magnitude, with a maximum conductivity when 40% (v/v) DMSO was added to the PEDOT:PSS. After this value, the conductivity starts to decrease, especially for concentrations >60%.
These results are in accordance with previous studies that have shown an increase in the conductivity of PEDOT:PSS when adding DMSO.[28,39,40] This phenomenon is probably due to the elimination of the excess of PSS/C0 from PEDOT:PSS.[34]

2.3. Effect of Multiple PEDOT:PSS Coatings on Conductivity of PEDOT:PSS Coated Yarns

To further increase the conductivity of the PEDOT:PSS coated yarns, multiple successive PEDOT:PSS coatings were applied. Figure 4b shows the variation in the conductivity versus the number of PEDOT:PSS coating layers at different DMSO concentrations. The conductivity, when no DMSO was used, resulted in the lowest conductivity and showed a linear increase in the number of PEDOT:PSS coating layers. Higher values of conductivity were observed when 40% and 80% (v/v) DMSO were used during PEDOT:PSS soaking, in accordance with the results shown in Figure 4a. In these cases, the conductivity increased up to four coatings, after which the conductivity remained constant (80% DMSO) or even decreased (40% DMSO).

Figure 4c shows the thickness of the PEDOT:PSS coated yarns after successive coatings. It can be observed that the thickness increased with the number of PEDOT:PSS coatings following a linear relationship. The weight also increased with the increasing number of coatings, which further indicates the presence of a thicker PEDOT:PSS coating (See Figure S1, Supporting Information).

2.4. Effect of the DMSO Concentration on the Actuation of the Textile Yarn

Once the conductivity of the yarns was ensured to be sufficient and optimized, PPy electropolymerization was performed on the PEDOT:PSS coated yarns to provide mechanical actuation.[7,41] The PPy synthesis was checked by visual inspection, SEM images, and EDX spectra (Figure 5). The presence of PPy coated on yarns can be clearly observed due to its darker black color (Figure 3c and 5b). The diameter of PPy-coated yarn in Figure 5b is higher than that of the PEDOT:PSS coated yarn in Figure 3c, which ratifies that PPy has been synthesized on the surface of the PEDOT:PSS coated yarn. Furthermore, the Figure 5b shows nodular/PPy cauliflower-like surface confirming the PPy addition.[42]

EDX spectroscopy (Figure 5c) shows the presence of O, C, S and Cu, in accordance with results shown in Figure 3d. In addition, N could be detected at 0.392 keV, which is an indication for the presence of PPy.[13]

The electroactivity of PEDOT:PSS coated yarn and PPy/EDOT:PSS coated yarn at different DMSO concentrations was investigated using cyclic voltammetry (CV). Figure 6a shows the CV responses of coated yarns measured at the 4th cycle in 0.1 M sodium dodecylbenzenesulfonate (NaDBS) aqueous electrolyte. A higher current through the PPy/PEDOT:PSS coated yarns can be observed as compared with the PEDOT:PSS coated yarn. Also, better defined redox peaks can be observed when PPy was present. This points to the fact that the electropolymerized PPy layer has better electroactive properties than the PEDOT:PSS layer obtained in the first coating, so reaction (1) takes place to a higher extent, doping and dedoping of the sodium cation also causing the elongation and contraction of the yarn.[43] Further comparison also shows that the 40% DMSO results in higher currents than using 80% DMSO, as would be expected from the conductivity data in Figure 4b.

Next, the exerted linear strain was measured (Figure 6b). For that, a digital microscope was used to record the movement of a clamp mounted at the end of the yarn (Figure 2b). The exerted strain of the PPy coated yarn during both oxidation and reduction reactions was observed by measuring the movement of the clamp during either oxidation or reduction reactions separately. Figure 6b shows the movement (contraction/expansion) of the PPy/EDOT:PSS coated yarn actuator. The movement up/down (contraction/expansion) during oxidation/reduction, respectively, confirms that the motion is driven by exchange of cations, according to electrochemical reaction (1).

The linear strain was calculated according to the following expression:[41]
Strain \( \% \) = \( \frac{\Delta L}{l_0} \cdot 100\% \) (2)

where \( \Delta L = l - l_0 \) is the length variation during oxidation/reduction and \( l_0 \) is the initial length of the CP-coated yarns.

Figure 6c shows the absolute values of maximum strain when the CP is oxidized or reduced.\(^{[41]}\) It is clearly observed that the PPy/PEDOT:PSS coated yarn has a larger actuation than PEDOT:PSS coated yarn pointing to the positive effect of the electrochemically polymerized PPy on the actuation. Furthermore, the actuation strain is higher in the yarns coated with the PEDOT:PSS/DMSO solution in both PEDOT:PSS and PPy/PEDOT:PSS coated yarns. A higher conductivity of the PEDOT:PSS coated yarns (Figure 5a) should promote a more uniform PPy electropolymerization as well as better oxidation and reduction of the polymerized PPy layer when using a high-conducting “current collector.”\(^{[44]}\) In addition to the linear change, it was also possible to observe a twisting movement to left/right during oxidation/reduction, respectively (See Figure S2, Supporting Information).

2.5. Effect of the Number of PEDOT:PSS Layer on the Yarn Actuation

The effect of the number of PEDOT:PSS layers at different DMSO concentrations on the actuation was investigated using CV. Figure 7a shows the CV responses of coated yarns measured at the 4th cycle in 0.1 M NaBDS aqueous electrolyte. Anodic and cathodic limits were varied in Figure 7a to be able to find the oxidation and reduction peaks. Higher currents were achieved in the PPy/(PEDOT:PSS)\(_6\) (PPy electropolymerized on a yarn with six consecutive layers of PEDOT:PSS coating) coated yarn than in PPy/PEDOT:PSS (PPy electropolymerized on a yarn with a single PEDOT:PSS coating) coated yarn. We attribute this difference to the higher conductivity from PPy/(PEDOT:PSS)\(_6\) layer (Figure 4b).

Figure 7b shows the absolute values of maximum strain when the CP is oxidized or reduced. It is observed that the PPy/(PEDOT:PSS)\(_6\) coated yarn presented a better actuation performance than PPy/PEDOT:PSS coated yarn for all (0%, 40%, and 80%) DMSO concentrations (v/v). Again, we attribute this difference to the higher conductivity of the (PEDOT:PSS)\(_6\) coated yarn, which results in a more uniform electropolymerization of PPy and better oxidation/reduction of the PPy layer. The difference is more evident at 40% DMSO.

Table S1, Supporting Information, summarizes all the calculated strain obtained from the different compositions of yarn. It also shows the oxidized or reduced strain values from different methods such as CV, chronomperometry, and chronopotentiometry. Chronomperometric experiments were performed by applying square potential waves from −1.0 to 0.5 V versus Ag/AgCl with a period of 200 s, whereas square current waves of ±0.1 mA (200 s period) were applied for chronopotentiometric experiments. Results concerning the strain values using chronomperometric and chronopotentiometric methods are also shown in Supporting Information (Figure S3 and S4). The maximum to minimum actuating strain is denoted as follows: PPy/(PEDOT:PSS)\(_6\) > PPy/PEDOT:PSS > PEDOT:PSS.

Figure 5. SEM and EDX images from coated yarns: a) Pictures of bare, PEDOT:PSS and PPy/PEDOT:PSS coated yarns, b) SEM image from a PPy/PEDOT:PSS coated yarn, scale bar 10 μm, c) EDX spectra from a PPy/PEDOT:PSS coated yarn. PPy/PEDOT:PSS coated yarns, being the PEDOT:PSS coating obtained using 1 coating.
for all the investigated yarns. It is observed that the strain is directly related with the conductivity from PEDOT:PSS. A high conductivity results in a uniform electropolymerization of the PPy layer which provide a larger actuation.

A comparison of the obtained strains and strains of previously reported yarn actuators based on traditional yarns combined with actuating technologies is listed in Table 1.

As shown in Table 1, the results obtained show a clear improvement over the strains obtained using CP-coated commercial yarns so far. In this work, we have doubled the strain as compared with previously reported using the same technology. Although the CP-based yarn actuator reported here did not achieve the strain values reported for other technologies, this CP yarn actuator technology does present other advantages such as...
Thermal expansion (i.e., heating the actuator) and hydrogel swelling (i.e., exposing to moisture) are not practical means of actuation for wearable actuators.

It is worth to mention as well that novel carbon nanotube yarns are being developed that use both thermal expansion and moisture swellable hydrogels as actuation means, with promising results.

3. Conclusion

We have demonstrated a new method to obtain textile yarn actuators comprising CPs and commercially available viscose-based yarns. The CP yarn actuators were prepared using a two-step coating method. First, the yarn was coated using a commercially available PEDOT:PSS dispersion with the additive DMSO to increase its conductivity and subsequently coated with PPy using electrochemical polymerization. The PEDOT:PSS layer forms an electrically conductive surface, allowing a well-controllable electrochemical polymerization of PPy to form the electroactive (actuating) layer. The conductivity and actuation performance of CP yarn actuators were increased by varying the concentration of DMSO and increasing the number of PEDOT:PSS layers. The obtained yarn actuators showed electrochemically induced movement having mechanical performance, strain, up to 0.630 ± 0.036% for a PPy/(PEDOT:PSS)_6/40%DMSO yarn better than previous reports of PPy-coated yarns. The results showed a fivefold better performance than previously developed yarn actuators.[7,41] However, it is worth investigating to use less harmful additives (replacing DMSO) in the future and revisiting this study. This study provides a simpler and more efficient alternative coating method toward soft, textile-based actuators for wearables and assistive devices. It opens a new way toward the interest in improving the uniformity, conductivity, and material composition of CPs coated yarns to amplify the strain. The knitting and/or weaving of those yarns into fabrics can also have synergetic effects of yarns.[6,7]

4. Experimental Section

Materials: DMSO and pyrrole were acquired from Sigma-Aldrich. Pyrrole was previously distilled under vacuum before use and stored at −20 °C. NaDBS was acquired from TCI Europe and PEDOT-PSS PH1000 from Heraeus Company.

Viscose ring spun 1 ply yarn (Hof Garn Farberi GmH, Germany) with yarn number Nm 60/1 (Ne 36/1) was used for this investigation and cut into different samples with 72.0 ± 0.2 mm length.

Methods: The solid-state conductivity measurements of the uncoated yarns were performed using an Multimetrix DMM220 digital multimeter. The resistance was measured over 14.00 ± 0.01 mm yarn length. The length of the yarns was obtained using a digital caliper (Mitutoyo Absolute AOS Digimatic) with a precision of ±10 μm and the thickness using a digital micrometer (Mitutoyo) with a precision of ±1 μm. Also, the conductivity was calculated using the following expression:[48]

$$\sigma = \frac{1}{\rho} = \frac{l}{A \cdot R} \quad (3)$$

Table 1. Comparison of the different yarn actuators.

| Yarn Actuating Technology | Maximum linear reported strain [%] | Reference |
|-------------------------|-----------------------------------|-----------|
| Viscose ring spun 1 ply yarn | Conducting polymer | 0.63 | This work |
| Elastane | Conducting polymer | 0.3 | [7] |
| Lyocell | Conducting polymer | 0.075 | [7] |
| Polyamide + carbon | Conducting polymer | 0.01 | [41] |
| Silicon + carbon | Conducting polymer | 0.12 | [41] |
| Polyamide + silver coated | Conducting polymer | 0.04 | [41] |
| Cellulose + carbon | Conducting polymer | 0.06 | [41] |
| Polyester + 1,2 × INOX 50 μm | Conducting polymer | 0.049 | [41] |
| Polyester + 1,2 × Cu/5n | Conducting polymer | 0.025 | [41] |
| Gold-coated polyester | Conducting polymer | 0.014 | [41] |
| Cotton yarn | Water-swellable hydrogel | 9 | [49] |
| Coiled polyethylene terephthalate (PET) | Thermal expansion | 8.9 | [50] |

Figure 7. a) CVs obtained from PPy polymerized on yarns with both a single PEDOT:PSS coating (PPy/PEDOT:PSS) and six consecutive layers of PEDOT (PPy/(PEDOT:PSS)_6) at different DMSO concentrations after four cycles in 0.1 M NaDBS aqueous electrolyte at 10 mV s⁻¹. b) Strain measurements from PPy/PEDOT:PSS and PPy/(PEDOT:PSS)_6 coated yarns obtained during CV shown in Figure 6a.
where $R$ is the resistance of a sample between two points along the yarn, $l$ is the length in cm, $A$ is the cross-sectional area in cm$^2$, $\rho$ is the resistivity, and $d$ is the conductivity in S cm$^{-1}$.

All the electrochemical experiments were performed in a single-compartment three-electrode electrochemical cell, connected to a potentiostat–galvanostat Autolab PGSTAT 204 from Metrohm Company, controlled by a personal computer through Nova 2.1 software.

The reference electrode was a BASI MF-2052 Ag/AgCl (3 mM NaCl) electrode. In this work, every potential is referenced to this reference electrode. Furthermore, all the experiments were performed at room temperature (22 °C).

The yarn actuators were constructed following a two-step coating process: first a thin layer of highly conducting PEDOT: PSS was deposited on the yarn by soaking a piece of yarn into a PEDOT: PSS dispersion containing DMSO at different concentrations to ensure sufficient conductivity in the yarn. The yarn was immersed (soaked) into PEDOT: PSS for 1 h and then dried at room temperature for 24 h (the yarn became dark blue) (Figure 2a). Then an active (activating) layer of PPY was deposited using electropolymerization to form an electroactive yarn (Figure 2a).

For electropolymerization, a cylindrical electrochemical cell with a diameter of 4 cm was used. The working electrode was set in the center of the cell, surrounded by a stainless-steel fabric acting as the counter electrode. The PPY coating was obtained in NaDBS (0.1 M) and 1 mM pyrrole (0.1 M) aqueous solution by applying a constant current of 0.5 mA through the yarns for 2500 s. The yarns were pre-tensed by hanging a plastic screw (cupper tape) to the electrical connection) before putting them into the SEM holder and attached to the SEM sample holder based using conducting double-sided cupper tape.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

actuators, conducting polymers, electromechanical strains, poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate), polypyrrole, textiles, yarns

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