Investigation of electrically conducting yarns for use in textile actuators

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Received 28 December 2017, revised 16 March 2018
Accepted for publication 29 March 2018
Published 5 June 2018

Abstract
Textile actuators are an emerging technology to develop biomimetic actuators with synergetic actuation. They are composed of a passive fabric coated with an electroactive polymer providing with mechanical motion. Here we used different conducting yarns (polyamide + carbon, silicon + carbon, polyamide + silver coated, cellulose + carbon, polyester + 2 × INOX 50 μm, polyester + 2 × Cu/Sn and polyester + gold coated) to develop such textile actuators. It was possible to coat them through direct electrochemical methods, which should provide with an easier and more cost-effective fabrication process. The conductivity and the electrochemical properties of the yarns were sufficient to allow the electropolymerization of the conducting polymer polypyrrole on the yarns. The electropolymerization was carried out and both the linear and angular the actuation of the yarns was investigated. These yarns may be incorporated into textile actuators for assistive prosthetic devices easier and cheaper to get and at the same time with good mechanical performance are envisaged.

Keywords: actuators, textiles, conducting polymers, textuators, conducting yarns

(Some figures may appear in colour only in the online journal)

Introduction

Industrial robots and automated devices are nowadays extensively used in industry, enabling large volume production of standardized commodities, saving costs and increasing the quality of the products [1, 2]. At the same time they are improved and becoming cheaper every day [3]. As such, robotic devices are becoming ubiquitous in humans’ daily life, as it is the case of robotic vacuum cleaners, lawn mowers or pool cleaners [4]. One field raising specific attention nowadays is assistive robotics: robots helping people with some disability or injury to perform their daily tasks or assisting the elderly [5, 6].

In order to interact with humans, end-users prefer that such assistive devices and robot assistants look, feel and behave in manners to which we are accustomed [7, 8]. Thus, the entire robot, its components and even the materials they are made of are being redesigned to provide new, more user-friendly experience for the end-user [9–11]. For instance, it would be advantageous if assistive robotic devices could be worn like items of clothing, more comfortable and pliable than current hard and rigid exoskeletons. Following this idea, our group recently developed textile actuators [12]. They combine a well-established technology (textile processing) with novel actuating materials, i.e. with conducting polymers (CPs) actuators. Mammalian muscles move after an ionic flow arriving to the muscle through the nerves, such ionic flow promotes a chemical reaction (ATP hydrolysis, with release of ions), and with it, the subsequent conformational changes of the muscle proteins and the muscle contraction [13]. The so-called artificial muscles based on CPs [14, 15] mimic
mammalian muscle functions [16]. However, there is still room to improve their performance. For instance, muscle cells are very well organized, all of them collaborating to achieve movement. On the other hand, CP chains part of the CP films constituting the CP actuators are poorly organized with regards to mechanical actuation [17–19]. The combination of CPs with textiles utilizes the same biomimetic principles where every CP coated yarn acts in a collaborative way to achieve an amplification of both force and strain [12]. The textile actuators were constructed from passive fabrics following a two-step polymerization process: first a thin layer of highly conducting poly(3,4-ethylenedioxythiophene) (PEDOT) was deposited through vapour phase polymerization to ensure sufficient conductivity of the fabric, then an active (Actuating) layer of polypyrrole (PPy) was electro-polymerized on top to produce the electroactive fabric [12]. The two layers were needed: in order to get large movements electropolymerized CP films have been proven to have better actuating properties than chemically synthesized CP films [20–22], but in order to carry out an electropolymerization a conducting surface is needed. However, such chemically polymerized conducting layer may result in a decrease of the actuating properties of the actuator: it can be too rigid counteracting the movement of the high electroactive second coating, especially when it is optimized to obtain high conductivity [23–25].

In this work, we omit the initial chemical polymerization step in the fabrication of textile actuators and investigate different conducting yarns and their potential as conducting yarns in textile actuators. The electropolymerization process, as well as the electro-chemo-mechanical performance of the PPy coated yarns is studied in order to identify the optimal electrically conducting yarn(s) for textile actuators. Using commercial electrically conducting yarns would enable an easier and more cost-effective fabrication of the textile actuators.

Methods

Sodium dodecylbenzenesulfonate (NaDBS) from TCI Europe was used as received. Pyrrole (from SAFC®, acquired through Sigma-Aldrich) was distilled under vacuum prior to use and stored at −20 °C. Ultrapure water was obtained from Milli-Q Plus water equipment and used for the experiments.

Polyamide + carbon (PA + C) from Jarden Applied Materials (USA) with carbon particles included in thermoplastic spinning material, silicon + carbon (Silicon + C) from LeMur S.r.l. (Italy) with carbon particles included in silicon spinning, polymide + silver coated (PA + Ag coated) from Imbut GmbH (Germany) with the yarn surface coated with Ag through electrolys, cellulose + carbon (Cellulose + C) from Thüringisches Institut für Textil-und Kunststoff-Forschung e.V. (Germany) with carbon particles included in the cellulose spinning, polyester + 2 × INOX 50 μm (PES + 2 × INOX) from ITP GmbH (Germany) with PES-yarn wound with INOX wire, polyester + 2 × Cu/Sn (PES + 2Cu/Sn) from ITP GmbH (Germany) with a PES yarn double wound with copper wire—the copper surface is coated with Sn and gold coated polyester (PES + Au coated) from Swicofil AG (Switzerland) with PES yarn plasma coated with Au yarns were provided by ITP GmbH, Gesellschaft für intelligente textile produkte, and cut into different samples with 43 ± 1 mm length.

The solid-state conductivity measurements of the uncoated yarns were performed using an Multimetrix® DMM220 digital multimeter. The resistance was measured over 38.2 ± 0.5 mm yarn length. The length of the yarns was obtained using a digital calliper (Mitutoyo Absolute AOS Digimatic) with a precision of ±10 μm and the thickness using a digital micrometer (Mitutoyo) with a precision of ±1 μm.

All the electrochemical experiments were performed in a single compartment three-electrode electrochemical cell, connected to a Compactstat potentiostat–galvanostat from Ivium Technologies (Eindhoven, The Netherlands) controlled by a personal computer through IviumSoft 2.756 [3] software. The reference electrode was a BASI MF-2052 Ag/AgCl (3 M KCl) electrode. Every potential in this work is referenced to this reference electrode. All the experiments were performed at 22 °C (room temperature).

For the electrogeneration, a cylindrical electrochemical cell with a diameter of 4 cm was used. The working electrode was set in the centre of the cell, surrounded by a stainless-steel fabric acting as the counter electrode. The polypyrrole (PPy) coating was obtained in 0.1 M NaDBS and 0.1 M pyrrole aqueous solution by applying a constant current of 0.5 mA through the yarns of the different materials for 1500 s. The yarns were pre-tensed hanging from the lower end a metallic clamp having a mass of 1.096 ± 0.098 g. The electrical contact between the conducting yarn and the clamp was avoided with an insulating sticky tape at the bottom of the yarn where the clamp was set. Those yarns that were not black prior to the electropolymerization, became black after PPy synthesis, indicating a proper PPy coating.

The movement of the yarns was analysed in a square shape electrochemical cell in 250 ml of 0.1 M NaDBS aqueous solution. The yarns were again pre-tensed using the same metallic clamp used for the electropolymerization. The movement was recorded using a Dino-Lite Edge digital microscope attached to a personal computer and controlled by DinoCapture 2.0 software (version 1.5.14.G).

The scanning electron microscopy (SEM) micrographs were taken using a Leo 1550 Gemini SEM operating at 4.00 KeV. The fibre related breaking force was obtained by Staatliches Prüfamt für das Textilgewerbe, Kulmbacher Str.76, 95213 Münchberg, who performed a tensile test following DIN EN ISO 2062. The fineness of the yarns was obtained without pre-tension.
Table 1. Solid state resistance (38.2 ± 0.5 mm of yarn length), conductivity and fibre related breaking force of the yarns.

| Yarn             | Composition                  | Rotated yarn | Resistance/Ω | Conductivity/S cm\(^{-1}\) | Fibre related breaking force/mN/tex |
|------------------|------------------------------|--------------|--------------|-----------------------------|-----------------------------------|
| PA + C           | Polyamide + carbon           | Yes          | 1.32 × 10\(^3\) ± 7 | 1.85 ± 0.29                | 352.8 ± 6.9                        |
| Silicon + C      | Silicon + carbon             | No           | 6.44 × 10\(^3\) ± 1.6 × 10\(^2\) | 1.20 ± 0.06                 | 4.0 ± 0.2                           |
| PA + Ag coated   | Silver coated polyamide      | Yes          | 2.14 ± 4.5 × 10\(^{-1}\) | 7.04 × 10\(^4\) ± 4.7 × 10\(^3\) | 191.0 ± 5.9                        |
| Cellulose + C    | Cellulose + carbon           | No           | 5.70 × 10\(^3\) ± 3.40 × 10\(^3\) | 6.74 ± 5.51                 | 74.0 ± 1.0                          |
| PES + 2 × INOX   | Polyester + stainless steel  | Wrapped yarn | 1.92 × 10 ± 6.8 | 2.44 × 10\(^3\) ± 6.3 × 10\(^2\) | 199.0 ± 9.4                        |
| PES + 2Cu/Sn     | Polyester + copper/tin       | Wrapped yarn | 2.20 × 10\(^{-1}\) ± 4 × 10\(^{-2}\) | 1.12 × 10\(^3\) ± 3.8 × 10\(^4\) | 71.9 ± 0.6                          |
| PES + Au coated  | Gold coated polyester        | Yes          | 7.93 × 10\(^2\) ± 7.33 × 10\(^2\) | 1.06 × 10\(^3\) ± 1.01 × 10\(^3\) | 343.5 ± 3.9                        |
yarns, when a current of 0.5 mA was passed through the yarns in 0.1 M NaDBS, 0.1 M Pyrrole aqueous solution at room temperature.

0.1 M NaDBS aqueous electrolyte at room temperature.

NaDBS aqueous electrolyte. Figure 1

to the PES

Figure 1 to the PES

Once we had ensured that the yarns conductivities are listed in table 1. The conductivity values performed on the bare yarns. The measured resistances and measured to ensure that the electropolymerization could be first, the resistance and the conductivity of the yarns was performed on the bare yarns. The measured resistances and conductivities are listed in table 1. The conductivity values’ range was very wide (five orders of magnitude), but all of them were higher than 1 S cm⁻¹, which is in the range of semiconductors, and high enough to allow the PPy electropolymerization on their surface.

Once the conductivity was ensured to be sufficient to perform a PPy electropolymerization on the yarns, we investigated possible reactions occurring on the bare yarns immersed in the electrolyte that could interfere with the electropolymerization process [26–28]. Thus, cyclic voltammograms up to ±0.3 V were obtained with the yarns in 0.1 M NaDBS aqueous electrolyte. Figure 1(a) shows that in the potential window of −1.3 to 1.5 V, only very low currents were passing through the yarns, indicating that no important electrochemical reactions were taking place for all the cases, except for the PES + 2 × Cu/Sn yarn, where the current passing through the electrode reached high values in this interval. Considering the low standard electrode potential of Cu [29], we believe that this high current could be attributed to Cu redox reactions. Therefore, electrosynthesis of PPy, which potential falls within this window should be possible.

Electropolymerization

Once we had ensured that the yarns (paying special attention to the PES + 2 × Cu/Sn yarn, as mentioned above) were suitable for PPy electropolymerization, pyrrole was added to the solution at a concentration of 0.1 M and a constant current of 0.5 mA was passed through the yarns in 0.1 M NaDBS, 0.1 M Pyrrole aqueous solution at room temperature.

Results and discussion

Suitability of the different yarns for direct PPy electropolymerization

First, the resistance and the conductivity of the yarns was measured to ensure that the electropolymerization could be performed on the bare yarns. The measured resistances and conductivities are listed in table 1. The conductivity values’ range was very wide (five orders of magnitude), but all of them were higher than 1 S cm⁻¹, which is in the range of semiconductors, and high enough to allow the PPy electropolymerization on their surface.

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Electropolymerization

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The PPy synthesis was checked by SEM. Figure 2 shows the SEM images of the different yarns before and after the PPy coating, both at the upper part of the yarn (1.0 cm near the electrical connection) and at the lower part of the yarn (4.5 cm from the electrical contact). The first differences that can be appreciated are the different constructions of the yarns: multifilament (case of PA + C, PA + Ag coated, Cellulose + C, PES + Au coated), monofilament...
coated on the surface of every single filament. Since the multifilament yarns comprise many individual, thin filaments, the effective surface area is much larger thus resulting in a thinner PPy coating, as the total amount of PPy should be constant.

### Electro-chemo-mechanical characterization of the yarns

The electroactivity of the PPy coated yarns was investigated using cyclic voltammetry (figure 3). The different yarns have different resistances. High resistance should give higher peak separation. Thus, in order to check the electroactivity of the deposited PPy potential limits and scan rates were adapted to be able to observe the PPy redox peaks as best as possible. In fact, the yarns having the highest resistances are PA + C, Silicon + C and Cellulose + C (figures 3(a), (b) and (d)), which correspond with the yarns where it is not possible to observe clearly the redox peaks. The electropolymerization resulted in a very different electroactivity (current passing through the electrode) depending on the core yarn.

In every case it is possible to identify at least either a PPy oxidation or reduction peaks or shoulders.

Studying the electrochemical behaviour through potential steps (figure 3(h)), analogous results could be observed: very different currents were passing through the electrode. In addition, some shoulders or even current maxima at longer times than the initial peak time can be observed. Such shoulders and maxima are again characteristics of the PPy redox reaction [40].

Simultaneously with the electrochemical experiments, the linear strain of the yarns was measured. This was done by recording the movement of a clamp mounted at the end of the yarns, used to add a mechanical tension, using a digital microscope (figure 4(a)). By comparing the positions of the clamp at the end of the reduction and the subsequent oxidation cycle it was possible to observe the lifting of the clamp (figure 4(b)), which corresponds to the shrinking of the PPy coated yarn. As can be seen the yarns shrink during the oxidation cycle and expand during the reduction cycle confirming that the motion is cation driven according to equation (1), when PPy is electropolymerized in the presence of a large, immobile counter ion (dodecylbenzenesulfonate, DBS−) and it exchanges cations with the electrolyte [41–43]:

\[
[(\text{PPy}^0)(\text{DBS}^-)_n(C^+)_m(S)_m] \rightleftharpoons [(\text{PPy}^{a+}) \times (\text{DBS}^-)_n] + n(C^+) + m(S) + n(e^-),
\]

where PPy0 represents the neutral polymer chains, DBS− are the macroanions trapped in the polymer during electropolymerization as explained previously. However, the thickness measurements to confirm this are not easy: the different yarns have different architectures, resulting in PPy being coated differently. PPy was coated on the surface of the filaments (both monofilament or multifilament yarns) or on the conducting part of the wrapped yarns. In addition, the thickness varied along the length of the yarn, which makes it even more difficult to compare. However, results do show a correlation: the highest PPy coating thickness corresponded to monofilament yarn (Silicon + C), followed by the wrapped yarns (PES + 2 × INOX and PES + 2 × Cu/Sn), where PPy was only coated on the surface of the conducting components, and thinner PPy on multifilament yarns, where PPy was coated on the surface of every single filament. Since the multifilament yarns comprise many individual, thin filaments, the effective surface area is much larger thus resulting in a thinner PPy coating, as the total amount of PPy should be constant.
Strain (%) = \frac{\Delta L}{L_0} \cdot 100\% , \tag{2}

where $\Delta L = l - l_0$ is the length variation due to CP shrinking/swelling during oxidation/reduction and $l_0$ the initial length of the PPy coated yarns. Considering strain variations while the CP is oxidized or reduced the maximum strain in absolute value was obtained (figure 4(d)). The strain values of the different yarns varied one order of magnitude: from the PPy coated PA + C or the PPy coated PES + Au yarns (both yarns present the highest fibre related breaking force, they are less stretchy, table 1) exhibiting a very low strain of $\sim 0.01\%$ to the PPy coated Silicon + C (both yarns present the lowest fibre related breaking force, they are more stretchy, table 1) yarn with a strain of 0.1\%. This indicates that the core yarn
has a major effect on the actuation performance. These measured strains are similar to those obtained from other CP coated passive yarns [12, 44, 45].

In addition, some twisting of the clamp (figure 4(e)), and thus yarn, could be observed for some yarns, with twisting angles up to 1° for the PPy coated PES + Au yarn. We attribute this effect to the pre-twisting which is present in some of the bare yarns, similar to twisting observed in the angular actuators using carbon nanotubes [46].

Based on this data we conclude that PPy coated Silicon + C or PPy coated Cellulose + C yarns are the best of the investigated core yarns for linear actuation, while PPy coated PES + Au yarn is interesting for angular displacement. Further optimization to, e.g. adjusting the potential ranges, is currently ongoing to improve their lifetime at maximum actuation [26, 27], as well as knitting and/or weaving those yarns into fabrics to further exploit the synergetic effects of yarns in textile actuators.

Since the textiles actuators are expected to be used as actuators in garments and wearables, they should be also resistant to external influences such as sweat during use or detergents during washing. Promising results have been found in this aspect regarding CPs: it has been shown that it is possible to obtain PEDOT coated yarns that can be washed [47] or that the inclusion of CPs nanoparticles improves the washability of paints [48, 49].

Conclusion

In this work we have investigated seven different kinds of conducting yarns to be used in yarn and textile actuators, in order to develop a simpler fabrication method for textile actuators. By employing conducting yarns, the initial chemical polymerization can be avoided and electropolymerization of PPy can be directly performed. When coated with the electroactive polymer PPy, all conducting yarns showed electrochemically induced movement having mechanical performance (strain) in the range of previously reported PPy coated yarns and in some cases (PPy coated Silicon + C)
with better performance than previously developed textile actuators. In a subsequent work we will optimize the synthesis and actuation conduction for PPY to further enhance the actuation performance as well as assemble the PPY coated conducting yarns into textile actuators. This study will allow to further develop textile actuators that can be easier and more cost effectively fabricated towards developing soft, textile based actuators for assistive devices, garments and wearables.

Acknowledgements

Authors thank Luise Böhme for her help with yarns and explaining their characteristics. This work was supported by Carl Tryggers Stifelsen (grant nr. CTS16:207), Swedish Research Council (grant nr. VR-2014-3079), the Erling-Persson Family Foundation (proj. 2017-10-09—Textile Actuators for Wearable Assistive Devices) and Promobilia Foundation (grant nr. F17603).

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