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Polypyrrole actuators working at 2–30 Hz

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Abstract

"Soft actuators" based on the conducting polymer polypyrrole (PPy) may be especially suitable for use in combination with human limbs. A research project under the European Union Quality of Life program (DRIFTS, Dynamically Responsive Intervention for Tremor Suppression, http://www.drifts.org/) focuses on the development of practical tremor suppression orthoses prototypes [M. Manto, M. Topping, M. Soede, J. Sanchez-Lacuesta, W. Harwin, J. Pons, J. Williams, S. Skaarup, L. Normie, IEEE Eng. Med. Biol. 22 (2003) 120]. One of the choices of actuation mechanism is to use conducting polymers.

The main challenge is to provide significant forces at the frequencies relevant to tremor in upper limbs: 2–16 Hz. Forces in the range of 0.1–1 kg are required. It has earlier been shown that utilizing the stiffness change instead of the length change may extend the useable maximum frequency by about a factor of 10 [J.D. Madden, R.A. Cush, T.S. Kanigan, I.W. Hunter, Solid State Ionics 113 (2000) 185]. The maximum frequency reached was, however, only about 1 Hz.

By optimizing the synthesis method, and the choice of counterion in PPy and in the electrolyte, a polypyrrole actuator able to yield significant force at up to 30 Hz has been made. The stiffness change turns out to be approximately 20 times faster than the change in length. Simple scaling up of the present data leads to a required total thickness of PPy (30 mm wide film) of 0.13 mm at 2 Hz, and 0.32 mm at 15 Hz for the 1 kg limit. The required mass of the actuator itself at 15 Hz is ∼100 mg. The results indicate the feasibility of using PPy actuators for tremor suppression.
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1. Introduction

Neurological tremor (i.e. involuntary oscillations) affecting the upper limbs is brought about by a variety of pathological conditions, presenting tremor of widely different amplitudes and frequency characteristics [1]. The DRIFTS project aims to create an experimental orthosis for suppressing upper-limb tremor. The idea is to create a wearable system to suppress tremor while preserving natural movement. The approach is to measure forces and accelerations in real-time and then to counteract the involuntary motion by means of a fast response actuation system. Several different types of actuators have been evaluated and developed: mechanical actuation, magneto rheological fluids and conducting polymers.

This paper describes our attempts to develop the conducting polymer actuator option. The main difficulty is the frequency range of interest to tremor, approximately 2–16 Hz. Conducting polymer actuators have been reported driven at even higher frequencies [2], but mostly using the bending bilayer response, which converts small strain values into large visible tip motions, but which does not involve the large forces needed for tremor suppression (corresponding to up to 1 kg). Linear actuators show decreasing strain values at higher frequencies with 0.007% reported at 30 Hz [3].

Polypyrrole (PPy) has been used to provide the required actuation response. The mechanism of actuation is the insertion/ejection of cations and anions as the polymer is redox cycled between the reduced and the oxidized states [4]. In addition, the reaction is complicated by the slower motion of a number of water molecules (up to ∼20 per mobile cation) in and out of the polymer caused by an osmotic process [5,6]. We have earlier proposed bypassing the slow solvent uptake part of the length change, and exploiting the change in the Young's mod-

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ulus instead [7], would improve the prospects for making an actuator with significant force at realistic tremor frequencies [8].

The present article presents the results obtained from further comparisons of the frequency dependence of the length change, and of the difference in measured stress proposed to be due to the change in stiffness between the oxidized and reduced states.

2. Experimental

PPy films with thicknesses between 10 and 50 μm were synthesized electrochemically by the galvanostatic method. Typical current densities were 0.1–1 mA/cm^2. Most of our original work was done using the anionic detergent dodecyl benzene sulphonate (DBS) in aqueous solution. Recently, however, better results with respect to speed of actuation have been obtained using the method of synthesis described by Kaneto and co-workers [9]. In this process, 0.25 M pyrrole and 0.50 M tetra-*n*-butyl-ammonium BF_4 ($TBABF_4$) were dissolved in methyl benzoate and polymerized at a current density of 0.2 mA/cm^2 on a polished titanium electrode.

The cycling experiments were performed in 0.5–1 M aqueous solutions of $NaBF_4$ or $NaPF_6$. The mechanical properties – length and stiffness change as functions of potential and actuation frequency – were measured using an electronic balance (Sartorius BP211 D) in combination with a translation stage (Physik Instrumente M-405.DG) for slow (<1–2 Hz) and precise measurements, and a load cell (Transducer Techniques GSO-25 or GSO-1K) in combination with a Steinmeyer LT105-50-LM translation stage for faster measurements.

The measurements were either performed by monitoring the length at a fixed force, or by monitoring the force required to obtain a constant length during cycling. The stiffness change is characterized by the difference in load (g) needed to maintain constant position. At low frequencies (less than ∼0.1 Hz) both the swelling and the stiffness change will contribute, but our results indicate that that the stiffness change dominates at the higher frequencies of relevance to the tremor application. Usually, each film was tested using several voltage ranges—ranging from a minimum equilibrium range of −0.25 to +0.76 V versus Ag/AgCl at slow speeds up to −6 to +6 V at the highest frequencies.

3. Results and discussion

3.1. Length change

Most of the early work on conducting polymer actuators put the emphasis on obtaining as large a relative length change as possible. The maximum obtained by us for the PPyDBS system was about 12% [10]. Fig. 1 shows the elongation as a function of actuation frequency for a $PPyBF_4$ polymer. The potential was stepped between −0.9 and +0.7 V. The load was 10 g, corresponding to a stress of 1.3 MPa. At 0.01 Hz, the effect of increasing the stress was investigated by measuring at 20 and 30 g loads (2.6 and 3.9 MPa). The elongation was the same within about 1%.

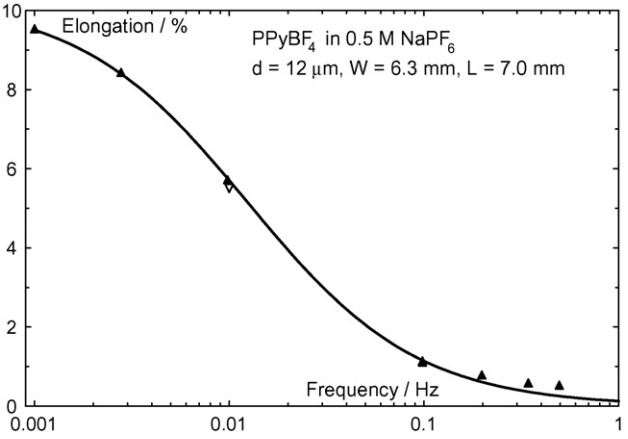

Fig. 1. Relative elongation difference between the oxidized and reduced states for a PPy film doped with BF_4^- as a function of the actuation frequency. Dimensions indicated on figure. The applied stress is 1.3 MPa.

The elongation is large (∼9.5%) at slow speeds, but drops off rather quickly: at 0.01 Hz only half is obtained, and at about 1 Hz, the effect is essentially non-existent.

This is below the desired range for tremor suppression and points to difficulties unless the inherent speed of the polymer film can be enhanced considerably (e.g. by using smaller dimensions to improve the speed of ionic diffusion). However, conducting polymers are not easily fabricated in the form of thin fibers.

3.2. Stiffness change

It is suggested in Ref. [8] that the stiffness change might be about 10 times faster. This proposition is tested in Fig. 2. The load at 0 V was 200 g, corresponding to 14 MPa.

The change in pulling force might be due to either the stiffness change, to a change in strain or to a combination of the two effects. We believe that under our experimental conditions, the stiffness change dominates above about 1 Hz, as evidenced by the comparison with the length change in Fig. 1. The results are

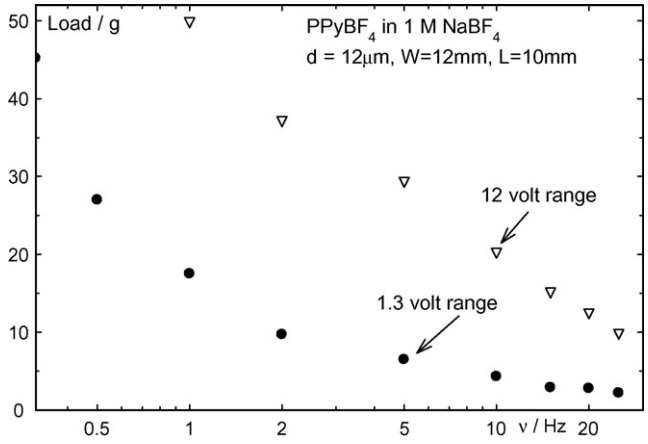

Fig. 2. The difference in load caused by the change in stiffness between the oxidized and reduced states of a $PPyBF_4$ polymer actuator. Dimensions indicated on figure. Results for two voltage ranges are shown. Ten grams corresponds to a stress difference of 0.68 MPa.

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