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High Force Polypyrrole Actuators

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Abstract

The force generated by a polypyrrole actuator operating under isometric conditions has been shown to decrease as the initial load applied to the actuator increases. The origin of the decrease in force generated was found to be the change in modulus that occurs upon electrochemical stimulation of the polypyrrole. A simple analytical model is introduced to describe these effects. Efforts to increase the force generated from polypyrrole involved the preparation of various bundles formed from hollow PPy tubes. Isotonic strains of approximately 0.5% were obtained at an external force of 2000 mN (5 MPa), higher than all previous reports.

Keywords

Polypyrrole, actuator, force generation

Introduction

Electromechanical actuators for practical use need to generate sufficient movement and sufficient forces to deliver the mechanical work required. Other considerations such as drive voltage are also vitally important, and there is a great interest in low voltage actuators, such as conducting polymers. These materials appear very attractive as actuators since they operate at <10 V and can generate strains of up to 12 %^[1] and at reasonable speeds^[2]. Much has also been made^[3] of the fact that the stress generated from conducting polymer actuators (~5 MPa^[4]) greatly exceeds that of natural muscle (~0.3 MPa^[5]).

Because of the relatively small size of the actuator materials developed to date, the actual displacements and forces are consequently small. A conducting polymer film of dimensions 10 mm x 1 mm x 0.05 mm producing an impressive strain of 10 % and a stress of 5 MPa only generates 1 mm movement and 250 mN force. Most applications at the macro scale require larger movements and higher forces. A “rehabilitation glove” for example capable of opening and closing the fingers on an adult hand requires 16mm movement and 1250 mN force^[6]. Some improvement in the movement generated from longer polypyrrole (PPy) actuators has been achieved recently by the incorporation of compliant metal electrodes^{[1] [2]} to reduce iR losses along the length of the actuator. Generating higher forces remains a challenge.

The actual force generated by an actuator depends upon its operating mode and is determined by the actuator strain produced, the material's Young's modulus and strength.

It has recently been noted that large shifts in Young's modulus occur during redox cycling of conducting polymers and influences the strain generated ^[7, 8]. Under isotonic conditions, the actuator strain generated is given by ^[8]:

$$\Delta L_f = \Delta L_0 - \frac{fL_0}{A} \left(\frac{1}{Y'} - \frac{1}{Y} \right) \quad (1)$$

Where ΔL_f is the actuator displacement generated at a constant force (f), ΔL_0 is the actuator displacement when the applied force is zero (the "free stroke"), Y is the Young's modulus in the expanded state, Y' is the Young's modulus in the contracted state, A is the cross-sectional area and L_0 is the initial length.

Under these conditions the amount of movement generated decreases with increasing loads since in all cases studied for conducting polymers $Y < Y'$. Under these conditions, the blocking force (that opposing force needed to prevent any movement) is predicted to be ^[9]:

$$f_{blocking} = \frac{\Delta L_0 AY}{L_0(\beta - 1)} \quad (2)$$

Where β is the ratio of Y/Y' . For the example listed above and where $Y = 100$ MPa and $Y' = 20$ MPa, $f_{blocking}$ is 250 mN. Very few reports are available describing the stress generated by conducting polymer actuators. Forces in the range of 100 mN - 300mN have been reported for polypyrrole films under isometric conditions ^[10-12]. Polythiophene gel samples have been shown to produce approximately 70 mN force in compression ^[13]. Other studies have considered the strain generated under isotonic conditions at different applied loads ^{[14] [8] [15]}. The maximum isotonic force that can be applied is limited to the

breaking strength of the actuator (typically < 10 MPa under dynamic conditions) so for typical cross-sectional areas, the maximum force achievable may be only 100 mN.

In some applications the actuator operates under isometric conditions and is required to generate a desired force during its operation. It is not known how the initial force (often required to straighten the actuator, especially when thin films or fibers are used) affects the final force produced. Experimental results and analysis of this situation are presented in this paper.

Finally, the often-touted strategy for producing high force, large movement and fast actuators from conducting polymers is to make bundles of parallel fibers (mimicking natural skeletal muscle). Some examples have already been reported ^[4, 16] but a full analysis of the effect of bundling conditions on actuator performance is lacking. This paper reports the study of small bundles of PPy fibers prepared in different ways, as illustrated in Figure 1.

Figure 1 a) Schematic diagrams showing assembly of bundled fibres; b) photographs of multiple strand bundles; and c) cross-sectional micrographs of single and triple bundled fibres.

Results and Discussion

Tests were conducted under both isotonic conditions at different applied loads to measure the actuator strain and under isometric conditions to measure the actuator force generated. In both tests the initial preload force was varied to determine its effect on the strain and force generated. Figure 2 shows typical results with the measurement of force and strain illustrated.

Figure 2

a) Typical results showing the displacement (strain) generated when the applied potential was $-1V$ under isotonic conditions; b) similar results for isometric testing where the change in force was measured.

Single Helix Tubes

Initially the performance of a single PPy helix tube was compared under isotonic and isometric conditions at different preload forces. Figure 3 shows the actuator strain determined under isotonic conditions at different (constant) tension forces. As has been described in several recent reports ^[8, 14, 15], the actuator strain decreases rapidly as the preload force increases. It is now well established that the isotonic strain is given by equation (1).

Figure 3

Actuator strain measured under isotonic conditions at different (constant) loads:

A: single helix tube

B: 4 helix tubes in parallel

The dashed line in Figure 3 represents the calculated results obtained using the fitting parameters given in Table 1. The apparent modulus value for the oxidized (Y) and reduced (Y') states are similar to those given in previous reports ^[7, 8]. The ratio of Y : Y' (β) is approximately 1.7, again similar to a previous report using a similar potential range. It is known that deep oxidation and reduction of polypyrrole films gives β values in the range 4 - 5. The low β values obtained in the present study are due to the relatively low applied voltages used (+/- 1 V) causing incomplete oxidation and reduction of the polymer.

Table 1

Samples, testing conditions and fitting parameters used for both isotonic and isometric analyses.

Sample	Condition	Free Stroke (%)	Free Force (mN)	Y (GPa)	Y' (GPa)	β
Single Fibre	Isotonic	4		0.075	0.045	1.7
Single Fibre	Isometric		140	0.075	0.038	2.0
4-Fibre	Isotonic	3.2		0.075	0.048	1.6
4-Fibre	Isometric		280	0.046	0.033	1.4

The isometric stress measured at different initial preload forces is given for the single helix fiber in Figure 4. The force generated when the voltage is applied decreases as the preload force increases, as would be expected from the decrease in actuator strain at higher preloads. The relationship between the force generated and the initial preload force can be determined by considering the 3 contributions to the total force separately. Firstly, the initial preload (f_i) causes an extension of the sample of ΔL_i which is estimated from (assuming linear elastic behaviour):

$$\Delta L_i = \frac{f_i L_0}{AY} \quad (3)$$

Figure 4

Actuator force generated under isometric conditions and at different initial applied loads:

A: Single helix tube B: 4 helix tubes in parallel

Upon application of a voltage that would cause shrinkage (typically from reduction) of the polymer under isometric conditions the force will increase by an amount Δf_0 . If it is assumed for the moment that the modulus does not change by application of that voltage,

then the polymer will attempt to shrink by an amount ΔL_0 – the free stroke, or the movement produced at zero preload. Equivalently, the free force (Δf_0) can be estimated from

$$\Delta f_0 = \frac{-AY\Delta L_0}{L_0} \quad (4)$$

Where if $\Delta L_0 < 0$ (contraction) then the force increases ($\Delta f_0 > 0$). However, it is known that the modulus of the polymer does change when a voltage is applied, and the change in modulus will lead to a further change in force (Δf_E):

$$\Delta f_E = \frac{A\Delta L_i}{L_0}(Y - Y') = \frac{f_i}{Y}(Y - Y') \quad (5)$$

The total force after application of the potential is then given by:

$$f_{total} = f_i + \Delta f_0 + \Delta f_E \quad (6)$$

And the change in force, or force generated (ignoring the initial preload force), from the application of a voltage is:

$$\Delta f_f = \frac{-AY\Delta L_0}{L_0} + \frac{f_i}{Y}(Y - Y') = \Delta f_0 + \frac{f_i}{Y}(Y - Y') \quad (7)$$

The dashed lines in Figure 4 are the values calculated from equation (7) and using the fitting parameters given in Table 1. In the isometric case, it is necessary to take a value of 2 for β which is slightly higher, but similar to the value used to explain the isotonic results. The value of Y has been determined so that the calculated free force (equation 4) is the same as the force generated when the data in Figure 4 are extrapolated to $f_i=0$.

The theoretical results shown in Figures 3 and 4 show excellent agreement with the measured values. It is therefore possible to calculate the force and strain generated from any given actuator as long as the input variables are known. It has previously been shown that different voltage ranges and different electrolytes affect the values of Y and Y' for polypyrrole ^[17]. In some circumstances (for example by using ionic liquid electrolytes) the difference in Y and Y' is very small. Thus, the force and stroke generated in these cases would change little at increasing preloads. The maximum force attainable would, therefore, be much greater.

Multiple Helix Tubes in Parallel

Figures 3 and 4 also show the isotonic strain and isometric force generated by a multi-fiber parallel assembly (sample B). The 4-fiber assembly shows the same strain at low loads, but higher strains at higher preloads than the single fiber. As seen from equation (1), the higher cross-sectional area (A) for the 4-fiber assembly accounts for the slower decrease in actuator strain at higher loads compared with the single fiber (small A). The dashed line in the figure shows the calculated results using the values for Y and Y' given in Table 1. Again, good agreement between theoretical and experimental results was observed and the fitting parameters were virtually identical for both the single PPy helix sample and the 4-fiber samples in parallel.

An additional advantage of the multi-parallel fiber assembly for high force applications is that it can sustain higher loads before failure than the single fiber. Again this is due to a higher cross-sectional area resulting in lower applied stress. In Figure 4, the highest

isotonic force applied was determined by the breaking strength of the material under active conditions. As seen from the Figure, the breaking force increases by a factor of ~ 4 , as expected from the four-fold increase in area.

The isometric force generated by the 4-fiber assembly at different tensioning loads is shown in Figure 4 as line B. Again, the force decreases as the preload force increases and as expected from equation (7). Careful comparison of the free force obtained from the 4-fiber assembly shows it to be approximately 3 times greater than that from the single fiber. A four-fold increase in free force would be expected from the difference in cross-sectional area (equation 4). Another small difference in expected behaviour assumed from simply a difference in cross-sectional area can be seen from the slightly different fitting parameters used for the single fiber and 4-fiber assembly under isometric conditions (Table 1). In particular, a much smaller modulus Y was needed to account for the measured free force for the 4-fiber assembly. The lower apparent modulus of the 4-fiber assembly reflects the difficulty in tensioning all fibers evenly in a multi-fiber assembly. If one fiber were tensioned less than the other fibers, then that fiber's contribution to the sample stiffness and actuation force would be reduced. The problems associated with achieving equal tensioning of all fibers within the system was found to be a major difficulty in producing high force actuators from parallel fiber.

Multi-Bundled Helix Tubes

A more reliable means for producing multi-helix assemblies, with all fibers in near equal tension, was therefore sought and two related methods are described in this section.

Firstly, a multi-helix bundle was formed by wrapping a platinum wire helically around two PPy coated helix tubes (sample C). Secondly, a platinum-wrapped double helix bundle was coated in PPy (sample D). Sample E was prepared by taking two sample D specimens, bundling and wrapping these with Pt wire then polymerising that bundle with PPy. Sample E therefore comprised 4 helix tubes, and a total of 7 PPy coated Pt helices.

The actuator strains measured under isotonic conditions for the multi-helix bundles are shown in Figure 5. The maximum strain obtained from these samples was slightly less than 2%, which is significantly lower than the single helix (~3%). The lower strain may be due to the additional PPy coated Pt helices that could restrict movement. The major benefit of the multi-helix bundles is seen in the higher loads that can be sustained – Sample E producing 0.4 % strain at preloads of 1000 mN. A similar 9-fiber sample was also constructed and found to give ~0.5% strain at 2000 mN. Previous reports have only demonstrated appreciable isotonic strains from PPy at loads of 250 mN^[15], 450 mN^[11] and 800 mN^[18].

Figure 5

Actuator strain measured under isotonic conditions at different (constant) applied loads:

C: two helix tubes wound with a Pt wire helix

D: as for A with an additional PPy coating

E: 2 double helix tubes wound with Pt helix and PPy coated (4 tubes, 7 helices)

Conclusions

The effect of applied tension on the actuator force from PPy has been reported for the first time. The change in force decreases with increasing preload tension as a result of the decrease in modulus that accompanies the contraction of the polymer (under the

conditions used). The total force (actuator generated force + preload force) increases with increasing preload. Applications that require a pre-determined force (eg. closing a clamp) are best performed at the highest preload tension that still allows a sufficient change in force upon application of the potential.

A simple analytical model has been proposed and verified to show how the actuator force varies with the preload tension under isometric conditions. The model applies equally to actuators operating in compression with differing applied loads. The model shows that higher forces are generated when the modulus shift is small. The type of actuator material and the operating environment have a significant effect on the modulus shift.

Finally, the preparation of high force polypyrrole actuators has progressed using the helix fiber as the basic unit. Parallel assemblies of helix fibers enabled more strain to be generated at higher forces (up to 600 mN), but difficulties were encountered in the equal tensioning of the fibers in the assembly. Alternative methods of bundling helix fibers and binding with additional PPy proved successful with the demonstration of significant actuation (0.5% strain) at a load of 2000 mN. This is the highest force reported to date for which appreciable strains can still be obtained from polypyrrole actuators.

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Experimental

The basic actuator structure studied was the “helix tube” as described previously ^[2]. The PPy helix tube consisted of a hollow fiber (internal diameter of 250 μm ; external diameter ~ 350 μm) of PPy, containing a thin platinum wire (diameter 50 μm) helix within the tube wall. The helix tubes were polymerised in a solution containing 0.05 M dopant (tetrabutyl ammonium hexafluorophosphate, TBA.PF₆) and 0.06 M monomer (pyrrole, PPy) in propylene carbonate (PC), using a current density of 0.1 mA cm⁻² for 24 hrs at 20°C. After polymerisation was completed the helix tubes were washed in PC, the inner wire removed and the tubes stored in an electrolyte solution, of 0.25M TBA.PF₆ in PC, until used.

The following samples were prepared for testing:

- A. a single PPy helix fiber was prepared by wrapping a 50 μm Pt helix around a 250 μm Pt wire and coating with PPy (1 fiber / 1 helix).
- B. 4 single PPy helix fibers (as in A) were placed in parallel and attached to each other only at the ends; not along the entire length of the fibers (4 fibers / 4 helices).
- C. 2 single PPy helix fibers (as in A) bundled with another external 50 μm platinum helix along the entire length of the fibers (2 fibers / 3 helices).
- D. 2 single helix tube substrates (uncoated) bundled with another external Pt wire along the entire length of the fibers, then all coated with PPy (2 fibers / 3 helices).
- E. 2 type D double helix fibers bundled together with another external Pt helix along the entire length of both fiber bundles, then all coated in PPy (4 fibers / 7 helices).

F. 3 helix tube substrates (uncoated) were bound with another Pt helix, then all coated in PPy (3 fibers / 4 helices).

A number of other bundled samples (eg. 9 helix bundles) were prepared and tested, but only the results from samples A-F are presented in this paper to provide a clearer insight into the effect of bundling on actuator performance. Figure 1 has a schematic diagram showing the assembly of the type E multi-helix bundle and a cross-sectional micrograph of samples A and F. The micrograph clearly shows the even coating that is produced over the single helix fiber and the hollow cavity suitable for carrying electrolyte. In the multiple helix sample F, the inner cavities were not coated in PPy because of the lack of penetration of electrolyte solution into the interstitial spaces.

Actuator testing was performed using the Lever Arm Dual Mode System (Aurora Scientific) that enabled forces up to ± 5 N and displacements up to ± 20 mm to be measured. Samples were firmly clamped at the lower end and the upper end connected to the lever arm by a stainless steel wire. The sample was fully immersed in electrolyte (0.25M TBA.PF₆ in PC) and connected to a GW Laboratory DC power supply (GPR_3030D). A PPy coated stainless steel mesh auxiliary electrode was used (that also acted as the reference electrode) in a two-electrode cell configuration. Figure 2 shows representative results taken under isotonic and isometric conditions, with the preload force and the actuator strain and actuator force defined. For the isometric testing the preload force was set for the polymer in its oxidized state. The contraction of the polymer during subsequent reduction then caused a further increase in force that was

measured as the “actuation force” and determined from the difference in force maximum and the preload force.

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