

Closed Loop Performance of Polypyrrole Linear Contractile Actuators

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Abstract—Conducting polymer actuators such as polypyrrole can generate stresses over 10 times larger than skeletal muscle and have typical repeatable strains between 1% and 12%, making them potential candidates for lightweight, low-cost, robotic applications. Polypyrrole linear actuators under closed loop control have not been previously reported. Here we report the open and closed loop performance of polypyrrole linear contractile actuators evaluated at pre-loaded stresses of 1 MPa to 3 MPa. A standard PI control scheme driving a potentiostat was implemented in conjunction with positioning feedback from a DC/DC linear variable differential transformer (LVDT). A dynamic positioning range of 3400 is reported, with a positioning resolution of 125 nm (0.001% strain) and a maximum repeatable displacement of 427 microns (3.6% strain). The open loop frequency response of actuator strain shows characteristics of a first-order low pass filter with a log gain versus log frequency slope near -1 for frequencies tested between 0.05 Hz to 2 Hz. The closed loop frequency response of actuator strain when tracking a sinusoidal set-point signal of 0.5% strain shows characteristics of a first order system with one zero, with a corner frequency near 0.08 Hz and an operating bandwidth up to 1 Hz. Step responses at various controller output maximum voltages show a reduction in contractile response times by a factor of four, where higher voltages yield faster contractile responses.

I. INTRODUCTION

THE conducting polymer polypyrrole can be used as a linear actuator when operated within an electrochemical cell. Because of its low voltage operation (1 V to 2 V), low-cost, relatively high force output [1], and ability to produce repeatable strains up to 12% [2], polypyrrole is seen as a potential candidate for artificial muscle applications in robotic, industrial and biomedical fields.

The application of polypyrrole actuators has been demonstrated in various real world devices. Polypyrrole's ability to operate in liquid environments makes it a potential candidate for use in maritime and aquatic devices. Polypyrrole fish fins mimicking the conformational shapes of the bluegill sunfish pectoral fin have demonstrated similar thrust patterns to that of the actual biological organism [3].

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A variable camber foil for use in ship propellers has been shown to provide adequate strain for modifying propeller shape underwater [4]. The use of polypyrrole in biomedical applications is also possible, where specific synthesis protocols can be modified to create bio-compatible materials that can actuate *in vivo* [5]. Polypyrrole actuators have also been used in a variety of miniature devices such as micro-positioners [6], pumps [7], and micro-grippers [8]. As performance, speed, force output and material characteristics continue to improve, the use of these actuators in closed loop robotic and micro-robotic applications becomes more and more viable.

Conducting polymer actuators are generally produced, tested, and integrated into systems in one of two configurations: bending actuators [9] and linear contractile films [2]. A design trade-off exists between these two configurations, with the former offering large displacements at the expense of lower force output and the latter offering high force output, but lower displacements. Both actuator configurations can suffer from relatively slow response times and creep damage over time. By incorporating feedback into polypyrrole actuators and limiting the working range of the actuator over its lifetime, significant improvements to minimize creep and increase speed can be attained. Closed loop feedback control can also offer precise output positioning for polypyrrole's use in a wider variety of applications.

To date, feedback performance studies on polypyrrole and other conducting polymers have been limited to bending configurations [10]. Closing the loop on bending polypyrrole actuators has been shown to both improve the response time of the polymer and reduce its positioning error [10]. Evaluating the linear contractile capabilities of polypyrrole within a closed loop system is of equal importance, especially in applications where high force output, precise positioning, or high actuator stiffness is mandated. Closed loop control also offers the possibility of programmable excitation waveforms, where relatively high voltage actuation can be exploited to reduce response times.

Relatively high voltage actuation (> 4 V) can increase the performance of closed loop polypyrrole actuators, but also reduce its operating life-cycle. Polypyrrole actuators operated at low voltages (< 1 V) and low pre-stresses (< 20 MPa) have shown strain reductions as little as 0.5% after 32,000 cycles [11]. At higher voltages and similarly low frequencies, this operating life-cycle is drastically reduced. At higher voltages (up to 10 V) and higher frequencies (above 1 Hz), however, 120,000 cycles of actuation at 0.25%

repeatable strain have been reported [12]. The trade-offs between voltage, frequency, and life-cycle are still being investigated to enable the long-term application of relatively high voltage actuation in closed loop polypyrrole actuation systems.

In this paper, the open and closed loop performance of linear contractile polypyrrole films are examined. Closed loop control is shown to significantly improve the positioning capabilities of the polymer films for both static and dynamic set-point targets. Relatively high voltage inputs (up to 9V) increase the polymer's actuation speed, but also reduce its operating life-cycle.

Although the samples studied were limited to operation at frequencies below 2 Hz, their large dynamic range of positions makes polypyrrole a feasible material for task-specific linear positioning devices where low-cost, high positioning accuracy is required.

II. INSTRUMENTATION AND PROTOCOLS

A. Polypyrrole Synthesis

Polypyrrole films were synthesized in a propylene carbonate solution of 1% (v/v) deionized water, 0.05 M tetraethyl ammonium hexafluorophosphate, and 0.05M pyrrole (Aldrich). Actuation tests were performed in neat 1-

butyl-3-methylimidazolium hexafluorophosphate (EMD Chemicals). All chemicals were used as received, with the exception of pyrrole which was distilled under vacuum before use.

Films were electrodeposited galvanostatically onto a glassy carbon crucible at a current density of 0.5 A/m^2 and a temperature of $-40.0 \text{ }^\circ\text{C}$. All films were grown for 8 hours, yielding thicknesses near 0.025 mm. After deposition, films were air-dried for 12 hours, sealed, and used within one week. Samples were cut to dimensions ($3 \text{ mm} \times 12 \text{ mm}$) and electrical contact was made via 0.030 mm gold wire and silver epoxy paste.

B. Instrumentation and Apparatus

Polypyrrole actuation tests were carried out in a custom built apparatus that allowed both electrochemical excitation of samples and continuous monitoring of both force and position data (Fig. 1). Film samples were held in place with miniature clamps machined from polytetrafluoroethylene. One clamp was statically fixed to a 45 N force sensor (LSB200, Futek) and mounted on the outside of the electrochemical cell. The force sensor was used to determine the initial pre-stress of the polymer under tension, and was excited using a signal conditioning amplifier (2311, Vishay). The other clamp was drawn by a 0.1 mm line of Spectra®

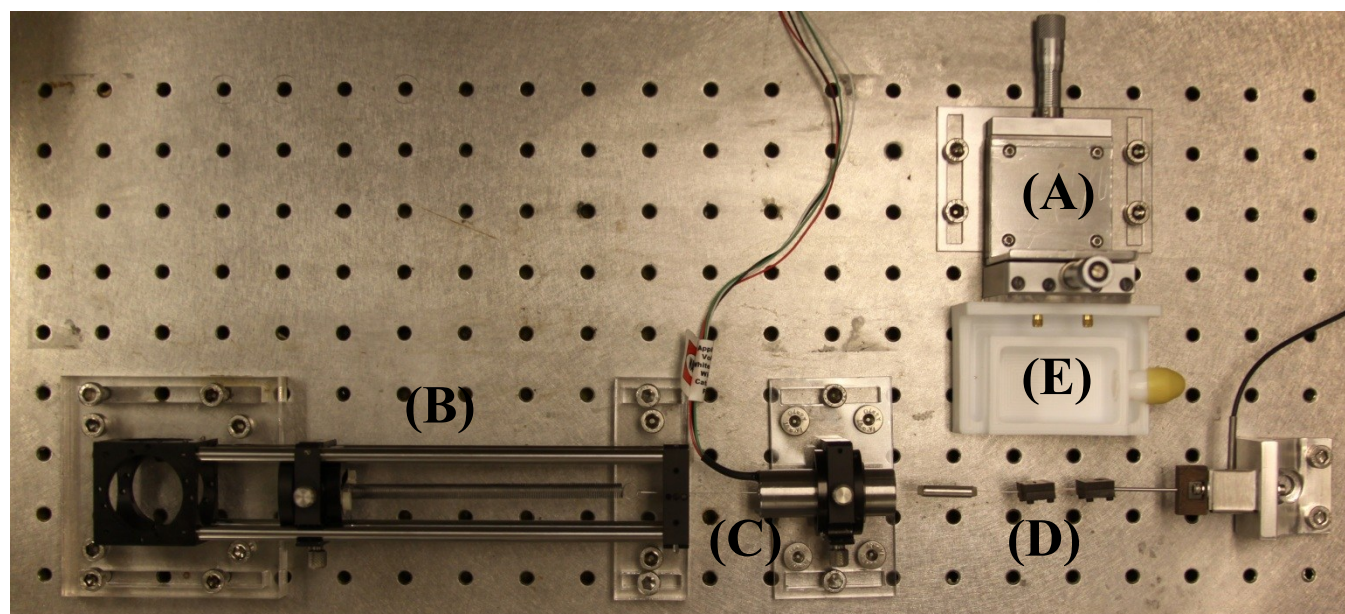
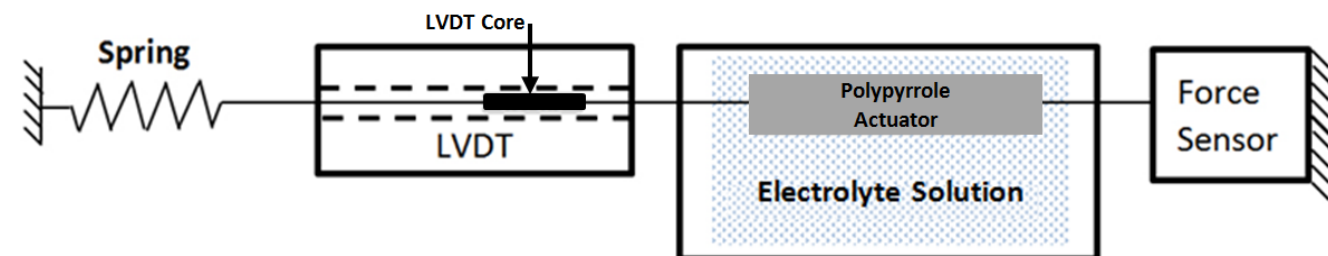


Fig. 1. Testing apparatus: The dashed lines (diagram) indicate hollow enclosures. Liquid levels in the bath were maintained above the maximum height of the sample. All components were fixed to an optical breadboard (below) to reduce vibration. The bath (E) and the outer casing of the LVDT (C) were mounted onto 2-axis positioning stages (A) or concentric optical mounts ensuring adjustments could be made to maintain concentricity along the line of action. The sample can be seen in-between the two clamps at (D). The total system is held under tension by the sliding spring (B).

cable and held under tension by a compliant extension spring (9640K123, McMaster-Carr). The force output from the polymer under contractile conditions was calculated to be at least two orders of magnitude higher than the reaction force exhibited by the spring for the given operating range of displacements that the polymer might undergo. The highly compliant spring thus provided a simple, low-cost, yet effective means of maintaining the polypyrrole linear actuator under quasi-isotonic tensile conditions.

A DC/DC linear variable differential transformer (LVDT) (75S1DC-050SR, Sentech) was axially aligned with the force sensor and the polymer film's contractile line of action. The LVDT core was mounted on the Spectra® cable and used to monitor displacements. Sag in the line from the weight of the LVDT core and the non-fixed clamp was minimized by keeping the clamp mass to a minimum and holding the line under tension.

An electrochemical bath consisting of a main chamber and secondary moat was used to test samples under horizontal tensile conditions. In the linear configuration, polypyrrole actuators were submerged in an ionic liquid during operation. Small holes on the outer walls of the chamber allowed the samples to be held under tension while the Spectra® cable itself never came into physical contact with the walls of the bath, thus eliminating any associated frictional impedances. In a full bath of ionic liquid, the holes on the outer walls were sufficiently small such that any liquid losses occurring over time were minimal compared to the size of the bath reservoir itself.

The main chamber was lined with 0.040 mm thick stainless steel foil which served as the counter electrode within the electrochemical cell. The reference electrode was a silver wire. The working electrode was the polypyrrole sample.

C. Excitation and Control Methods

The diffusion of ions in and out of the polymer leads to the film expanding or contracting volumetrically. This change can be exploited to generate forces or produce motion. Experiments on conducting polymers are usually performed under isotonic or isometric conditions. Under isotonic conditions [13], [14], [15], the polymer film is held fixed at a constant stress, is excited using a potential input, and the strain is independently measured. Under isometric conditions [16], [17], the length of the actuator is maintained constant and the stress output is measured during excitation. For real world applications, isotonic conditions can be approached by using springs or counterweights.

The polypyrrole actuator was driven using a potentiostat. In a standard three electrode configuration, a DC or AC command voltage was input into the potentiostat (2051, Amel Instruments). The potentiostat then drove a current between the counter electrode and the working electrode, such that the potential difference between the working electrode and the reference electrode equaled the input command voltage. As the potential across the actuator changed, the polymer was driven through cycles of oxidation and reduction, leading to the diffusion of counterions from the surrounding ionic solution [14] into or out of the

polymer film. This flux of ions yielded actuator displacements.

Control was performed using a real time program developed in LABVIEW®. For the basic control scheme (Fig. 2), the LVDT signal served as the process variable. An output voltage ranging between +/- 10 V was input into the potentiostat to drive the polypyrrole actuator. For the majority of tests, the controller output was limited to +/- 0.75 V with respect to the resting potential of the electrochemical cell, which ranged between 0.20 V and 0.45 V. The LVDT had a ripple voltage of 200 mV, which could be attributed to excitation of the primary and built in signal conditioning circuitry. The ripple voltage frequency (above 4 kHz) was several orders of magnitude higher than the actuation frequencies tested, and was therefore filtered out in post processing. Although the ripple voltage did not allow real-time visual confirmation of tracking performance at low positioning increments, its relatively high frequency with respect to the system response time meant its overall effects on system accuracy were minimal, as they were averaged over time.

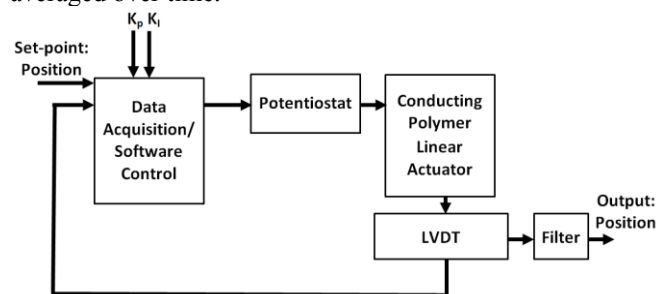


Fig. 2. Control scheme for driving polypyrrole linear actuators. Voltage signals input into the potentiostat drive the polypyrrole actuator between cycles of contraction and expansion. Data acquisition and software control were performed on a computer running Windows XP. A National Instruments PCI Data acquisition card (NI-6289) was used for both acquiring and generating signals.

For all tests, proportional and integral control were used. PI gains were calculated with aid of an auto-tuning algorithm built into LABVIEW® and were maintained constant for the data presented at: $P=1466$, $I=0.049$. AC and DC set-point signals were computed in real-time and in buffered configurations.

III. OPEN LOOP CONTROL

Open loop control of polypyrrole linear actuators was evaluated by exciting the polymer with a discrete set of sinusoidal inputs between 0.005 Hz and 2 Hz, with a constant amplitude limited to +/- 0.75 V with respect to the resting potential of the electrochemical cell. The output strain was sampled at 100 Hz for each frequency input. A single sine wave fit was computed on each set of strain measurements for the frequencies tested. Actuator strain output, frequency and phase information were calculated from each fit using a custom script written in MATLAB®. The resultant Bode plot can be seen in Fig. 3.

Open loop performance of the actuator's strain response is similar to the open loop electrical response of polypyrrole previously studied [18]. Similar response characteristics are

expected since strain is roughly proportional to charge. Without feedback, the system shows the characteristics of a first-order low-pass filter for the frequencies tested, with a log gain versus log frequency slope of -1. At low frequencies the phase lag is reduced, suggesting that ions have more time to move in and out of the actuator and the response is dominated by electrical and ionic resistance. At frequencies near 0.1 Hz, diffusion becomes the rate limiting phenomenon and the phase remains fixed near -55 degrees.

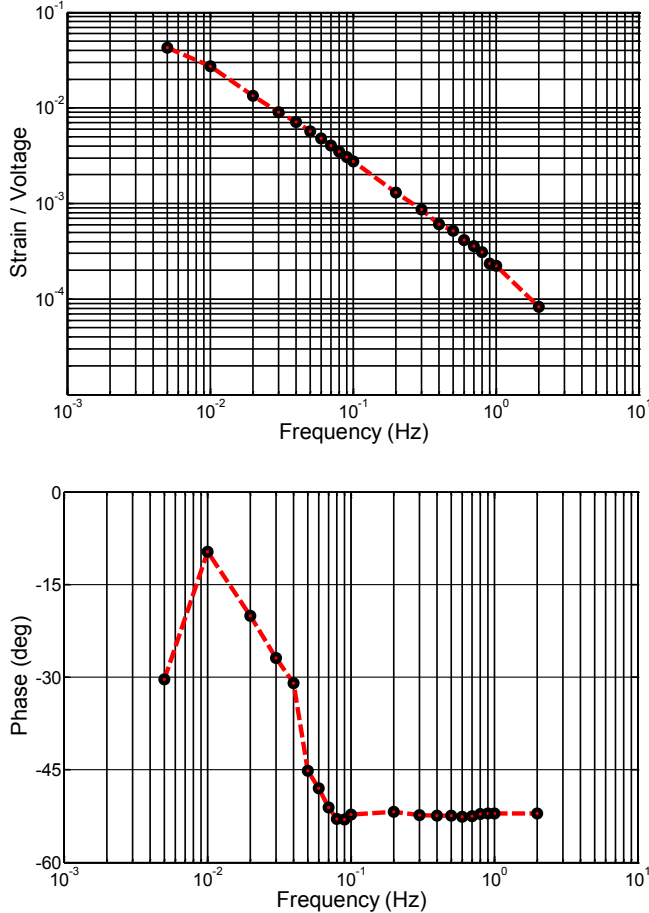


Fig. 3. Open loop response of strain per volt (ϵ/V) and phase for linear contractile polypyrrole. The data points represent the discrete frequencies tested. The line was drawn using nearest neighbor interpolation.

Because polypyrrole actuation is a diffusion limited process, output strain decreases for a given input amplitude as the frequency increases. This can be attributed to the fact that there is less time for ions to enter and leave the polymer. Faster responses can be achieved by increasing the potentiostat's maximum output voltage [12], decreasing the polymer's thickness dimension, or modifying the deposition protocol to increase conductivity [2], [19]. Under such modifications, the curve in Fig. 3 would shift to the right. Studies are still undergoing to see if such modifications would enable moderate strains above 1 Hz to be achieved.

Open loop control of conducting polymer actuators can be used in situations where oscillation or relative motion is required, rather than absolute positioning. Because the polymer exhibits creep over time [11], without feedback

there is no way of maintaining steady positions over long periods of time. Fig. 4 shows one example of an open loop strain response to a 0.03 Hz, ± 0.75 V sine wave. The phase offset between the two signals has been removed and the outputs have been normalized to compare the shapes of the waveforms. The general shape of the output strain matches the input for the low frequency being examined, but under an open loop configuration, polymer creep becomes an issue. Creep is observed in Fig. 4 only over a few cycles. Over the lifetime of the polymer, errors between the input and output may increase dramatically.

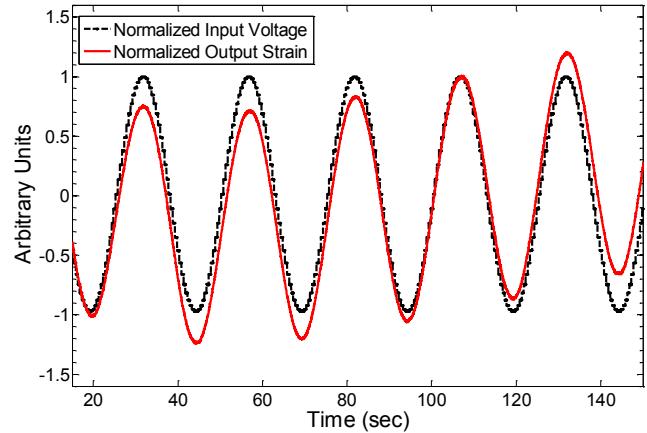


Fig. 4. Open loop response to a sinusoidal input, 0.03Hz, ± 0.75 V, 0.219 V offset (cell resting potential). The phase shift has been removed and the curves were normalized for comparison. The y-axis is both a measure of normalized voltage and normalized strain. The output response shows a similar shape to that of the input, but creep is apparent in the increasing mean of the output strain.

IV. CLOSED LOOP CONTROL

A. Frequency Response

Closed loop performance was evaluated in both the frequency and time domains by incorporating the LVDT as positioning feedback within a PI control scheme.

A sinusoidal signal of 0.5% strain was used as a set-point target. The maximum controller output ranged from ± 0.75 V with respect to the resting potential of the cell. The actuator's strain was measured using the same sampling and fit parameters as described in Section III. The resultant Bode plot is shown in Fig. 5. The closed loop system response has characteristics of a $1/(s + \tau)$ integral response, with unity gain at low frequencies, a corner frequency near 0.08 Hz, and a slope near -1 for frequencies above 0.1 Hz.

Like the open loop configuration, modifications made to the polymer's composition, dimensions, or maximum controller output would increase the actuator's response capabilities and shift the curve to the right. For smaller target set-points, the usable operating range would also increase.

B. Non-periodic Set-point Tracking

An arbitrary waveform was also used as a set-point signal to evaluate the strain response over time for non-periodic signals. In Fig. 6 a series of intermittent triangular and DC signals were randomly created to generate an arbitrary set-

point target. The total signal output was adjusted so that the target maximum strain remained below 0.5%. The actuator output plot shows the strain following the waveform. Because the system is comparably slow (on the order of seconds), the actuator has time to reach the set-point and maintain tracking. Long-term tracking of DC signals (data not shown) can also be performed under a closed loop configuration.

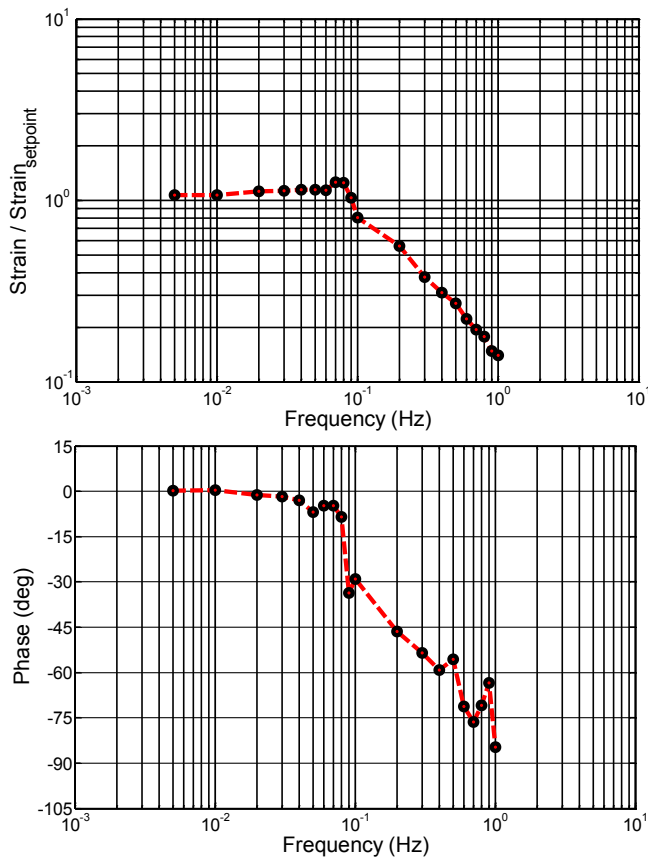


Fig. 5. Closed loop response strain (δ) normalized about the set-point. The data points represent the discrete frequencies tested. The line was drawn using nearest neighbor interpolation.

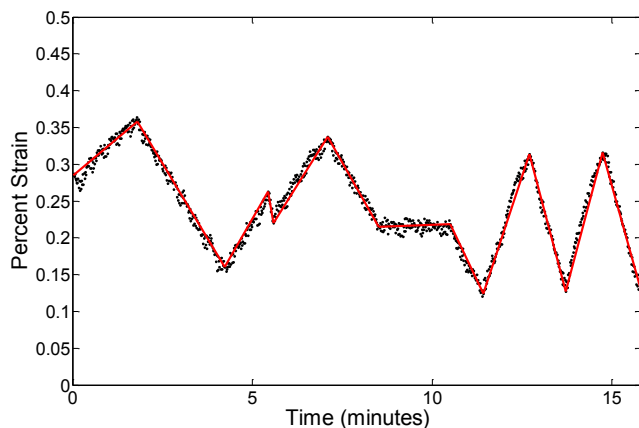


Fig. 6. Arbitrary waveform tracking composed of a series of triangular inputs of various slopes and intermittent sections of DC values. Strain was limited to 0.5% over the total operating range. Potentiostat current was limited to 1 A. The initial strain offset has been removed.

C. Incremental Positioning Resolution

Polypyrrole's closed loop positioning resolution was evaluated by setting the target set-point to a series of 125 nm incremental steps, spaced over 60 second intervals. Data were collected at 1 kHz. Post-process filtering on the LVDT positioning signal was performed using a moving window smoothing average with a window size equal to that of the sampling rate. A small section of steps is shown in Fig. 7.

The incremental response shows both the controller and the polymer actuator are capable of maintaining high positioning resolution where absolute accuracy is important. The step size of 125 nm corresponds to a strain of 0.001%.

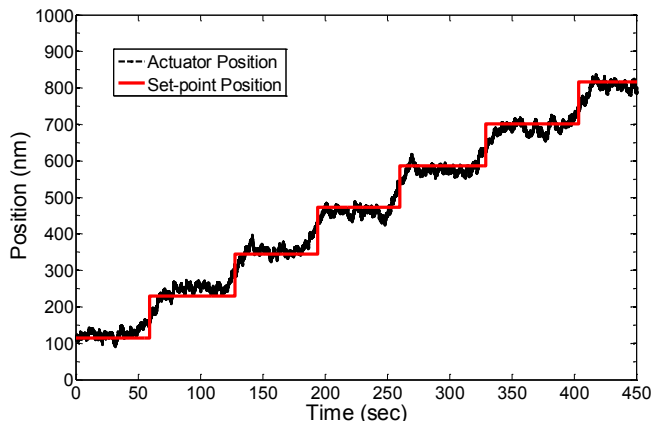


Fig. 7. 125 nm step increments under closed loop control. Set-point and actuator response agreement suggests that high positioning resolution or, slow incremental strains can be generated.

With a maximum displacement of 427 microns (3.6% strain) for the same sample tested at low frequencies, the polypyrrole actuator tested has a dynamic range near 3400. Studies are still ongoing to determine whether or not this resolution is length dependent or is limited by the feedback and processing of the system.

D. Step Responses

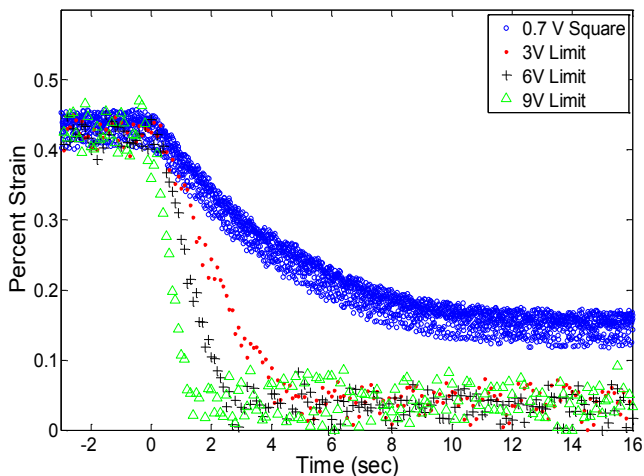


Fig. 8. Polypyrrole closed loop step response at different maximum controller outputs. 0.7 V square is the open loop response. Initial strain offsets have been removed.

The response to a fixed set-point step at various maximum

controller voltages was also evaluated to benchmark performance gains in response times of the actuators. Previous studies have shown that relatively high voltage inputs yield fast actuator responses, but at the expense of polymer degradation [12]. Fig. 8 shows the step response of the closed loop system at various maximum controller outputs. For higher maximum controller outputs, the response time is reduced by a factor of 4 between 3 V and 9 V. The difference in responses suggests that the system is not slew-rate limited by the controller itself, but that a non-linearity is introduced by the actuation mechanism of polypyrrole.

V. DISCUSSION

The performance of open and closed loop polypyrrole linear contractile actuators yields several results that can be used as reference when designing or incorporating conducting polymer actuators into robotic applications. The open loop response can be used in part to predict system behavior for the frequency and voltage range tested. Long oscillation times will yield larger strains. As the frequency of the actuator approaches 1 Hz, strain output becomes less useful for any application that requires the actuator to perform work at moderate strains.

In the closed loop configuration, a working bandwidth can be predicted in two ways. First, one can choose a target strain anywhere between the maximum and minimum values of the open loop system. For a smaller target strain, the bandwidth will increase, but at the expense of a limited strain range. Second, one can increase the controller voltage output. This will yield faster actuator response times and therefore a wider bandwidth. Modifying deposition protocols, changing the actuator dimensions, and working with different ionic solutions will also shift the bandwidth in one direction or the other.

Appropriate controller design for polypyrrole actuators will depend on the desired application. For applications where speed is critical, advanced controller design could be exploited to increase the performance metrics in a closed loop system while reducing the temporal exposure of the polypyrrole actuator to higher voltages. For applications where speed is not critical or relatively static set-points exist, an adaptive controller might operate under normal or even lower voltage ranges to conserve power.

The dynamic range of polypyrrole and its ability to output a controllable strain in small increments are useful features for low-cost, precise positioning applications. Whether the positioning resolution of polypyrrole actuators is a function of controller hardware is still being investigated. If it turns out that, like actuator strain, the lower limit of incremental strain is length independent, then scaled versions of linear polypyrrole contractile actuators will have the best of both worlds: larger total displacements without sacrificing the step size.

REFERENCES

- [1] Madden, P.G.A., Madden, J.D.W., Anquetil, P.A., Vandesteeg, N.A. and Hunter, I.W., "The relation of conducting polymer actuator material properties to performance," *IEEE Journal of Oceanic Engineering*, vol. 29, no. 3, pp. 696-705, July 2004.
- [2] Pytel, R.Z., Thomas, E.L., Chen, Y. and Hunter, I.W., "Anisotropic Actuation of Mechanically Textured Polypyrrole Films," *Polymer*, vol. 49, no. 5, pp. 1338-1349, January 2008.
- [3] Tangorra, J.L., Anquetil, P.A., Fofonoff, T., Chen, A.Y., Del Zio, M. and Hunter, I.W., "The Application of Conducting Polymers to a Biorobotic Fin Propulsor," *Bioinspiration and Biomimetics*, vol. 2, June 2007.
- [4] Madden J.D.W., Schmid, B., Hechinger, M., Lafontaine, S.R., Madden, P.G.A., Hover, F.S., Kimball, R. and Hunter, I.W., "Application of Polypyrrole Actuators: Feasibility of Variable Camber Foils," *IEEE Journal of Oceanic Engineering* 2004, vol. 29, no. 3, July 2004.
- [5] Smela, E., "Conjugated Polymer Actuators for Biomedical Applications," *Advanced Materials*, vol. 15, no. 6, pp. 481-494, March 2003.
- [6] Smela, E., Kallenbach, M. and Holdenried, J., "Electrochemically Driven Polypyrrole Bilayers for Moving and Positioning Bulk Micromachined Silicon Plates," *J. Microelectromechanical Systems*, vol. 8, no. 4, pp. 373-383, 1999.
- [7] Wu, Y., Zhou, D., Spinks, G.M., Innis, P.C., Megill, W.M. and Wallace, G.G., "A Conducting Polymer Based Microfluidic Pump," *Smart Materials and Structures*, vol. 14, no. 6, 2005.
- [8] Smela, E., Inganäs, O., Pei, Q. and Lundström, I., "Electrochemical muscles: micromachining fingers and corkscrews," *Advanced Materials*, vol.5, pp. 630-632, April 1993.
- [9] Takashima, W., Pandey, S. and Kaneto, K., "Investigation of bi-ionic contribution for the enhancement of bending actuation in polypyrrole film," *Sensors and Actuators B*, vol. 89, no. 1-2, pp. 48-52, March 2003.
- [10] Yao, Q., Alici, G. and Spinks, G.M., "Feedback control of tri-layer polymer actuators to improve their positioning ability and speed of response," *Sensors and Actuators A: Physical*, vol. 144, Issue 1, pp. 176-184, May 2008.
- [11] Madden, J.D., Rinderknecht, D., Anquetil, P.A. and Hunter, I.W., "Creep and cycle life in polypyrrole actuators," *Sensors and Actuators A: Physical*, vol. 133, Issue 1, pp. 210-217, January 2007.
- [12] Madden, J.D., Cush, R.A., Kanigan, T.S. and Hunter, I.W., "Fast contracting polypyrrole actuators," *Synthetic Metals*, vol. 113, no. 12, pp. 185-192, June 2000.
- [13] Spinks, G.M., Truong, V.T., "Work-per-cycle analysis for electromechanical actuators," *Sensors and Actuators A: Physical*, vol. 119, no. 2, December 2005.
- [14] Vandesteeg, N., "Synthesis and characterization of conducting polymer actuators," Ph.D. dissertation, Dept. of Materials Science and Engineering, Massachusetts Institute of Technology, Cambridge, MA 2007.
- [15] Pytel R., Thomas E. and Hunter I., "Anisotropy of Electroactive Strain in Highly Stretched Polypyrrole Actuators," *Chemistry of Materials*, vol. 18, no. 4, 2006.
- [16] Kaneto, K., Fujisue, H., Kunifusa, M. and Takashima, W., "Work behaviors of artificial muscle based on cation driven polypyrrole," *Smart Materials and Structures*, vol. 16, no. 2, June 2007.
- [17] Pytel, R.Z., Thomas, E.L. and Hunter, I.W., "In situ observation of dynamic elastic modulus in polypyrrole actuators," *Polymer*, vol. 49, pp. 2008-2013, January 2008.
- [18] Madden, P.G., Madden, J.D. and Hunter, I.W., "Parallel Electrochemical Methods to Accelerate Electroactive Material Discovery and Optimization," *Mat. Res. Soc. Symp. Proc.*, vol. 698, 2002.
- [19] Fofonoff, T., "Fabrication and Use of Conducting Polymer Linear Actuators," Ph.D. dissertation, Dept. of Mechanical Engineering, Massachusetts Institute of Technology, Cambridge, MA 2008.