REPORT DOCUMENTATION PAGE	*Form Approved* *OMB No. 0704-0188*

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1. REPORT DATE *(DD-MM-YYYY)*
2/23/12

2. REPORT TYPE
Final Report

3. DATES COVERED *(From - To)*
3/1/2009-11/30/2011

4. TITLE AND SUBTITLE
NASTIC ACTUATION: ELECTROOSMOTIC PUMPING FOR SHAPE-CHANGING MATERIALS

5a. CONTRACT NUMBER

5b. GRANT NUMBER
FA9550-09-1-0125

5c. PROGRAM ELEMENT NUMBER

6. AUTHOR(S)
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5d. PROJECT NUMBER

5e. TASK NUMBER

5f. WORK UNIT NUMBER

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8. PERFORMING ORGANIZATION REPORT NUMBER
Final Report

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AF Office of Scientific Research
Arlington, VA 22203

10. SPONSOR/MONITOR'S ACRONYM(S)

11. SPONSOR/MONITOR'S REPORT NUMBER(S)
AFRL-OSR-VA-TR-2012-0356

12. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for Public Release

13. SUPPLEMENTARY NOTES

14. ABSTRACT
The goal was to demonstrate a flexible actuation technology that would enable smart materials with both large stresses and strains. Using microchannels that would pump fluid by electroosmosis to deform flexible materials, we experimentally demonstrated devices that created large material deformations at high pressure, and whose performance matched theoretical predictions.

15. SUBJECT TERMS

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT	b. ABSTRACT	c. THIS PAGE			
					19b. TELEPHONE NUMBER *(Include area code)*

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Nastic Actuation: Electroosmotic Pumping for Shape-Changing Materials

Shapiro, Smela, Fourkas

Introduction and Background

We had developed a new type of polymer actuator based on electroosmotic pumping of fluid from one place to another within an elastomeric material. Theoretical calculations showed that these actuators should be able to deliver both high stress and high strain. The goals of this research were to perform the basic research required to achieve that goal, which would lead to applications such as soft robots, color-changing materials, and micro-manipulators. The research topics included the reduction of electrolysis of the pumping fluid, optimization of the channels to achieve high force, and design of the fluidics to achieve desired shape changes in the material. In the first year, our research focused on the first two.

Results Summary

Our first nastic prototype consisted of a supply chamber connected to an expansion chamber by a number of microchannels. The elastomer was polydimethylsiloxane (PDMS) and the pumping fluid was water. A high voltage (3.5 kV) was applied between platinum wire electrodes separated by 3 cm (E = 0.1 V/μm), resulting in reversible membrane deflections of ~300 μm within approximately 10 seconds. We had identified hydrolysis as the main factor limiting greater force output and began researching methods to reduce these electrochemical reactions, we have increased the actuation pressure, and also made important advances in fabrication/ integration. The electroosmotic pressure can be increased by reducing the channel size. The nastic actuator technology was therefore developed to incorporate porous polymer monoliths into the microchannels to achieve high-pressure pumping. We devised methods for producing unit cells that pump horizontally as well as cells that pump vertically, which can be combined into more complex structures to create actuations in different directions and for different shapes (e.g. to enable soft robots working in confined spaces). Initial multi-unit prototypes, actuated pneumatically, have been produced and modeled. Components required for the robots, such as compliant electrodes, have also been fabricated. A key aspect to be addressed in developing the nastic actuator is bubble formation within the microchannels due to hydrolysis of the pumping fluid. Research toward bubble-free actuation was performed, and although this remains a challenge, there are promising approaches that are being tested, including pumping alternate fluids.

Increasing Force/Pressure

The maximum pressure P that can be achieved by the actuator is given by $\mathrm{P} = \dfrac{12\varepsilon\varepsilon_0\zeta\left|\vec{\mathrm{E}}\right|}{\mathrm{d}^2\left[1-0.63\mathrm{d/w}\right]}$, where ζ is the zeta potential, E is the electric field, d is the channel diameter, and w is the channel width. The zeta potential depends on the surface chemistry, and can be positive or negative, which determines the direction the fluid moves. Increasing ζ results in a linear increase in P. A high electric field can be maintained while lowering the voltage by reducing the electrode spacing. Since the electrochemistry at the electrode/fluid interfaces depends on the voltage, this is important to address; this is also discussed below. The biggest impact on the force is obtained by reducing the channel size because it comes in as the square.

Porous polymer monoliths (PPMs) consist of a solid phase filled with small pores. They were originally developed for lab-on-a-chip devices, with applications in chromatography [1-5] and solid phase extraction [6-8]. They have also been used for electroosmotic pumping [9, 10]. Over the last two years, our group developed a method to produce porous polymer monoliths of porous poly (BuMA-co-EDMA) within PDMS microchannels (Figure 1a), defining the location of the monolith within the channel using UV light of 365 nm [11]. These monoliths are well anchored to the channel walls and can withstand high pressure.

Figure 1. a) Cross-sectional SEM image of a porous polymer monolith in a PDMS microchannel. b) PPMs (white) within the channels of a nastic device. c) Pressure increase due to PPM.

The PPMs have been integrated within a nastic actuator (Figure 1b), and the pressure with and without the PPM was measured using a commercial sensor (Figure 1c). *The pressure increased significantly*, but not by the expected orders of magnitude that have already been demonstrated by other groups [10, 12, 13]. This is because the pressure was limited by the formation of gas by hydrolysis of water at the electrodes due to the high applied voltage (kV), not by the performance of the monolith. (In the previous work [10, 12, 13], the systems were open, not sealed, so this was not an issue.) Thus, the challenge of reducing or eliminating the electrochemical reactions must be addressed before the monoliths can be utilized.

Reducing Electrolysis

The most significant risk in realizing this technology is the electrochemical reactions within the fluid cells. In water, hydrolysis produces hydrogen and oxygen gas:

$$2H_2O(l) + 2e^- \rightarrow H_2(g) + 2OH^-(aq) \quad \text{and} \quad 4OH^-(aq) \rightarrow O_2(g) + 2H_2O(l) + 4e^-$$

The generation of gas between two electrodes separated by a gap is illustrated in Figure 2a in two parallel channels. In addition to lowering the actuation pressure, gas bubbles block the current flow causing pressure fluctuations, and when a bubble completely fills the channel, the actuator ceases to function. The performance of the devices changes as the pH of the pumping fluid is altered by the creation of OH^-. In addition, the pumping fluid is depleted as the gas forms and diffuses out through the device walls.

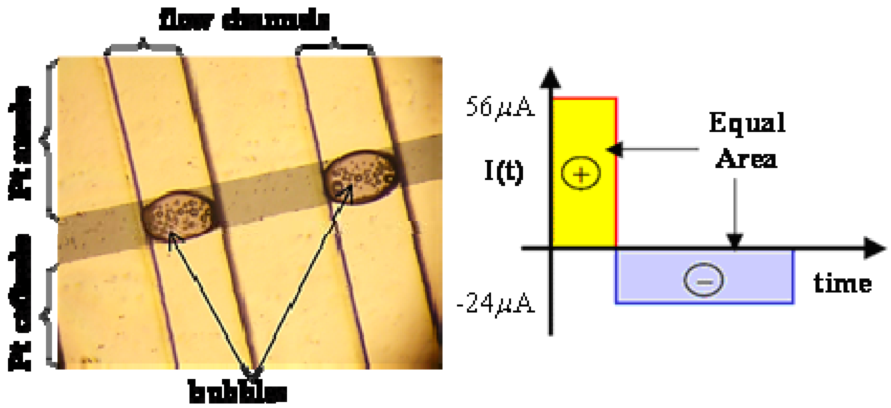


Figure 2. a) Bubbles forming within microchannels. b) Biased AC field driving concept with zero-averaged current input.

Literature reports suggested that electroosmosis can be achieved using asymmetric AC fields, instead of a DC field, based on the motion of tracer particles [14-18]. We tested this method, applying a biased zero-averaged asymmetric current signal (Figure 2b) at low frequency. When the movement of microbeads was recorded for different duty cycles, the particle velocity increased with duty cycle. Furthermore, no bubbles formed. Unfortunately, *there was no associated fluid movement*, as determined by measuring the fluid at the channel outlets. This suggested that the tracer particles were moving under electrophoresis, rather than being carried by EO-induced fluid motion.

Integration/Fabrication

Compliant Electrodes For the soft robot to deform, the electrodes must be able to stretch while maintaining electrical conductivity. We have taken two approaches in our group, developing composite compliant electrodes consisting of exfoliated graphite nanoparticles mixed into an elastomeric host [19], and corrugated thin film metal electrodes [20]. The former can be applied by spray- or spin-coating. Both have been patterned onto the elastomer. We used SU-8 photoresist as a mask to pattern the metal, resulting in crack-free films (due to swelling of the PDMS, etching and lift-off result in cracks).

Cell Filling External tubing has been eliminated in the prototypes. The device in Figure 3c was filled using a 30 gauge fine needle to inject the fluid. The elastomer re-sealed around the perforation, and the device could be operated without any perceptible leakage.

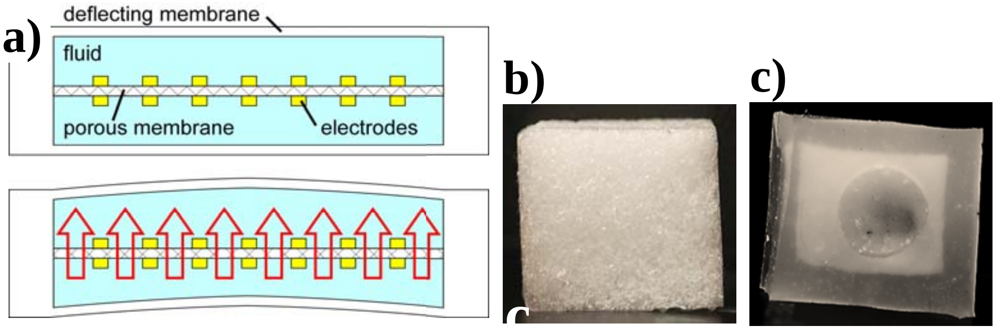


Figure 3. a) Schematic of unit cell with porous elastomer membrane for pumping vertically. b) Porous elastomer c) embedded in unit cell device.

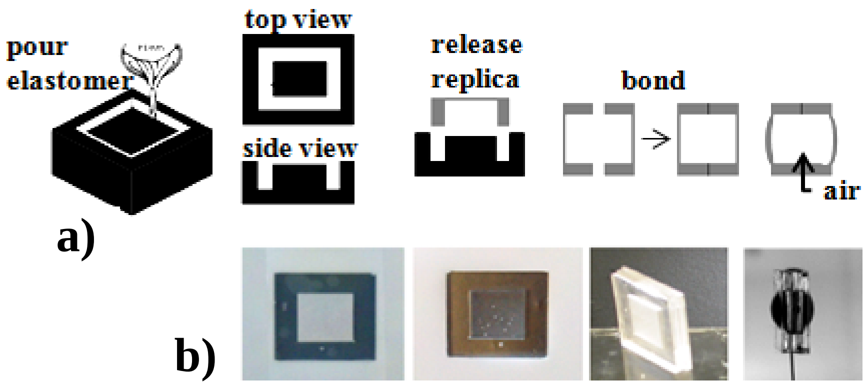


Figure 4. a) Fabrication of unit cells with high aspect ratio having membrane-thin deflecting walls. Laser machining is used to create the molds. b) Two pieces are bonded together to form each cell. The final panel shows an air-filled cell bulging outwards.

Cell Fabrication To allow arbitrary motion of the robot, cells are needed that expand/contract within the plane (pumping right-left) as well as cells that expand/contract out of the plane (pumping up-down) (Figure 3a). Our previous prototypes pumped within the plane, having horizontally-oriented channels (Figure 1a). In the last year, we have also created a porous membrane (Figure 3b), similar in concept to the PPM, that can be placed between fluid chambers. A high-aspect ratio process is required to create a tall device with very thin membranes (Figure 4). Using laser-machining we have been able to make molds that produce structures with a 1:100 aspect ratio. We have demonstrated that this configuration pumps successfully.

Design Membrane deflection was predicted by performing a bending analysis simulation. The actuator cell was modeled as being fixed to a cantilever beam (the flexible membrane), and a moment balance was solved over a range of deflections to find the membrane pressure required to bend the actuator to a certain angle (Figure 5b). Experimental results from a cell that was pneumatically inflated to known pressure (Figure 5a) were comparable to those predicted by the model (Figure 5c).

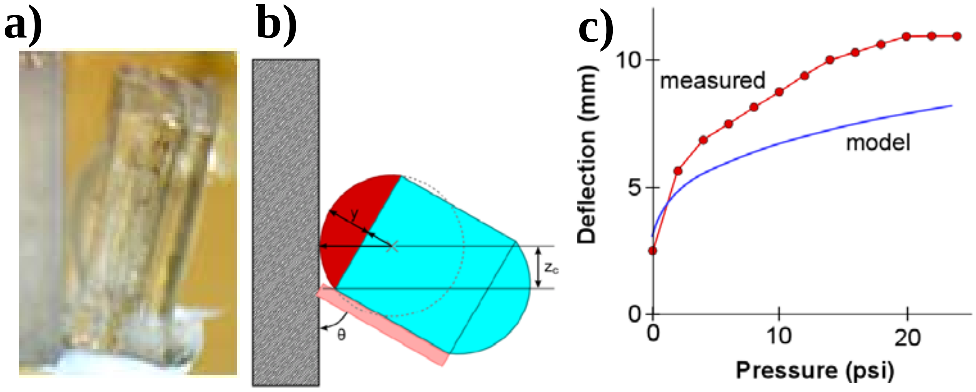
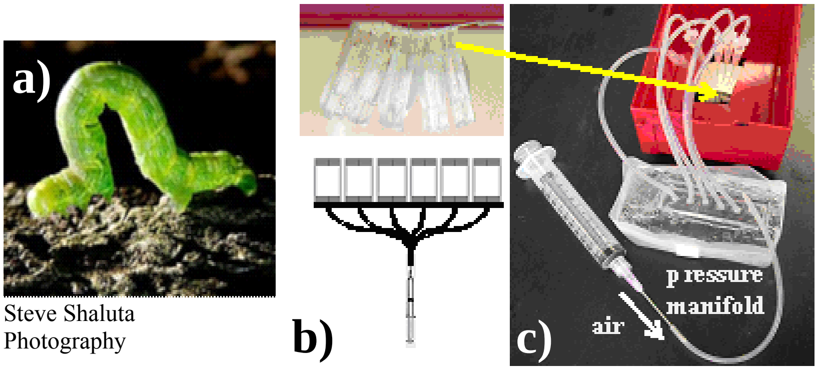


Figure 5.
a) Pneumatic cell on flexible membrane, deflecting as membrane bulges.
b) Model.
c) Deflection vs. Pressure.

Inchworm Prototype Pneumatic actuation was used for prototyping our first multi-cell inchworm-like structure (Figure 6), constructed by placing the vertically-pumping cells adjacent to each other on an a flexible sheet. The actuators are arranged such that membrane bulging occurs along the horizontal direction and each actuator displaces the neighboring actuator, resulting in bending the sheet. Such an arrangement will be used to realize inchworm-like motion. The next step will be to actuate these units using electro-osmotic flow.



Steve Shaluta Photography

Figure 6. a) Inchworm. b) Pneumatically-actuated series of cells mounted on a thin bendable sheet and c) the pressure manifold.

The electroosmotically-actuated cells do not require external tubing.

Smaller Channels and Fabrication Methods for Mass Production

The Fourkas group has been able to successfully make silica prototype devices that were derived from master structures fabricated from our specially formulated SU-8 using high-speed multiphoton absorption polymerization (MAP). The silica prototypes were created in collaboration with Prof. Shoji Maruo of Yokohama National University. The devices are functional, and the embedded channels allow for fluid flow within the fused silica. The prototypes were made from a single SU-8 master structure. This technique gives us the means of mass-producing microfluidic systems with high solvent compatibility and readily adjustable surface chemistry.

We have also been successful in the fabrication of arrays of polydimethylsiloxane (PDMS) channels with high surface area (aspect ratios up to 20:1) connected to reservoirs. We have developed methods for functionalizing the interior walls of different devices to alter the zeta potential for electroosmotic flow applications.

We are currently working on increasing the density of channels in these devices by decreasing the diameters of the channels to < 5 micron and increasing the numbers of channels in each device. We are also working to fabricate devices containing channels with heights greater than 1 mm. The robust physical properties of our specially formulated SU-8 allow for this type master structure to be replicated with high fidelity. The next step will be to embed this channel array into silica using our silica molding process. This type of device will be able to withstand the pressures of such a high surface area network.

Conclusion

Nastic's micro-channel pumping to create material actuators that achieve large deformations at high pressures (high stress and strain) was demonstrated experimentally. From a first year demonstration, the performance of the system has been improved and higher forces/pressures have been demonstrated using thinner channels, polymer monoliths and flexible electrodes. Limiting effects, such as electrolysis, have been reduced, and methods are being developed to enable mass production of such flexible smart actuation materials.

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