July 8th, 2012

Dr. Peter Gregory
Editor, Advanced Materials
Postfach 10 11 61
69451 Weinheim
Germany

Title: “Soft Machines That are Resistant to Puncture, and That Self Seal”

Authors: Robert F. Shepherd, Adam A. Stokes, Rui M.D. Nunes, and George M. Whitesides

Institution: Harvard University
Department of Chemistry and Chemical Biology

Figures: 3
Dear Peter,

We attach a copy of this manuscript for consideration as a Communication in *Advanced Materials*. This manuscript has not appeared in or been submitted to any other journal.

Soft Robotics is a field that uses materials and structures able to deform at low stresses (typically 100’s of kPa) to build machines that interact safely with their environment and perform sophisticated functions with relatively simple control systems. We are using organic chemistry, and materials science, to increase the functionality of these robots. For these systems, our material of choice in fabrication has primarily been silicone-based organic elastomers.

Silicone actuators enable large strains at low actuation pressures; however, they are susceptible to puncture from sharp objects (e.g., thorns). When punctured, they leak pressurized air, and their pneumatic actuation mechanism is rendered ineffective. This property limits their use outside of the laboratory. In this paper, we fabricate actuators using a stretchable composite of elastomer and fibers in a pleated geometry that reduces their sensitivity to puncture, makes them self-sealing if they are punctured, but also retains a range of motion similar to that of the purely elastomeric actuators. The addition of fibers also prevented local weakening of the elastomer during repeated actuation, a process that typically led to failure in previous, purely elastomeric Pneu-Nets.

The availability of a material platform that is soft, tough, and insensitive to puncture is a significant step towards the use of soft robotics in unpredictable and damaging environments.

Best Regards,

George M. Whitesides
We suggest the following individuals as possible referees:

Prof. Ilhan Aksay  
Chemical and Biological Engineering Department  
Princeton  
Engineering Quadrangle, Room A326  
Olden Street  
Princeton, NJ 08540  
Tel: (609) 258-4394  
Fax: (609) 258-6835  
Email: iaksay@princeton.edu

Prof. Annette Hosoi  
Department of Mechanical Engineering  
Massachusetts Institute of Technology  
77 Massachusetts Avenue, 3-262  
Cambridge, MA 61801  
Tel: (617) 253-4337  
Fax: (617) 253-7952  
Email: peko@mit.edu

Prof. Heinrich Jaeger  
James Franck Institute, E229  
University of Chicago  
929 East 57th. St.  
Chicago, IL 60637  
Tel: (773) 702-6074  
Email: h-jaeger@uchicago.edu

Prof. Daniela Rus  
Computer Science and Artificial Intelligence Laboratory  
Massachusetts Institute of Technology  
77 Massachusetts Avenue, 32-374  
Cambridge, MA 02139  
Tel: (617) 258-7567  
Fax: (617) 253-6849  
Email: rus@csail.mit.edu

Prof. Barry Trimmer  
Department of Biology  
Tufts University  
200 Boston Ave., Suite 2600  
Medford, MA 02155  
Tel: (617) 627-3924  
Fax: (617) 627-0909  
Email: barry.trimmer@tufts.edu

Prof. Manoj Chaudhury  
Department of Chemical Engineering  
Lehigh University  
111 Research Drive, Iacocca Hall  
Bethlehem, PA 18015  
Tel: (610) 758-4471  
Email: mkc4@lehigh.edu
Soft Machines That are Resistant to Puncture, and That Self Seal

Robert F. Shepherd¹, Adam A. Stokes¹, Rui M.D. Nunes¹, George M. Whitesides¹,²*

¹ Department of Chemistry and Chemical Biology, Harvard University,
12 Oxford Street, Cambridge, Massachusetts 02138

² Wyss Institute for Biologically Inspired Engineering
60 Oxford Street, Cambridge, Massachusetts 02138

*Corresponding author, email: gwhitesides@gmwgroup.harvard.edu
Abstract

This manuscript describes a soft gripper that is resistant to—and self-sealing after—puncture. This new capability in soft machines is enabled by embedding fibers in pneumatic elastomeric actuators to prevent the propagation of tears. These fibers reduce the extensibility of the actuators; using designs incorporating pleating compensated for this decrease, and allowed useful amplitudes of actuation of these systems. This pleated architecture had an additional effect of allowing these actuators two directions of movement: positive curvature under positive pressure, and negative curvature under vacuum. To demonstrate the functionality of these actuators, a soft gripper was made that was resistant to puncture and that could grip fragile objects (e.g., a wine glass) using both convex and concave gripping modes.

Introduction

We are developing a new class of actuators, machines, and robots—fabricated largely in organic elastomers—that are soft and compliant.[1-5] These systems require fewer components than hard robots, and can generate surprisingly complex motions based on their ability to deform at low stresses.[6-13] These machines are simple to operate, are light weight, can have a low center of gravity, and are low cost (relative to more familiar hard machines—that is those fabricated in metals).[1,2,4,5,14] We anticipate one application of these machines to be in operations involving environments too dangerous for humans (e.g., search and rescue, or the exploration of sites that may be toxic, dangerous, unstable, or radioactive).

We use pneumatic actuation, based on the anisotropic expansion of elastomeric chambers connected by networks of mm-scale channels (Pneu-Nets).[1,3,4] These polymeric structures are highly strained during actuation; at these strains, certain designs for the polymeric structures thin, and tear easily. When punctured, they fail; and even without external damage, repeated
actuation of purely elastomeric Pneu-Nets eventually leads to weakening, bulging of the pneumatic channel, and rupture of the internal walls—what we call an “aneurysm”—and to pressure-driven tearing and failure. To build tougher (the material property that describes how much energy our device can absorb without mechanical damage)\textsuperscript{[15]} soft robots, we need new materials and mechanical designs for our actuators.

Fibrous materials of both natural (e.g., cellulosic) and synthetic (e.g., polyaramid) origin can have elastic moduli in the range of gigaPascals; these materials are, however, typically not highly extensible, and not immediately compatible with the designs we have used for elastomeric Pneu-Nets. There are a number of designs that circumvent this limitation to Pneu-Net-based actuation. For example, by manually folding paper, we developed bellows-like actuators (e.g., pleated structures, as in the folds of an accordion) composed of intrinsically inextensible material to form extrinsically extensible structures.\textsuperscript{[16]}

The folds in the inextensible, fibrous material that comprise bellows are typically sewn and they prescribe the controlled collapse and expansion of the system; at the laboratory scale, however, sewing or other forms of joining are too labor-intensive for rapidly iterating soft robot designs. To mold an exact replica of a bellows in a single step (one that is folded on all sides) would require sacrificing a piece of our mold for each actuator.

\textbf{Results}

\textit{Materials & Methods}
This paper describes a bellows-like structure that can be fabricated by molding, without sacrificing parts of the mold; unlike true bellows architectures, the materials we use must have some intrinsic extensibility (~100% strain before failure to achieve actuation amplitudes similar to those of the Pneu-Nets described previously;[1,4] Fig. S1).

We fabricated these actuators from a composite material: polyaramid fibers (Fig. S2 shows a representative fiber bundle) embedded in an elastomeric matrix. They operate, in part, by the folding and unfolding of a quasi-bellows arrangement of pleats. This design brings four advantages over our un-pleated, purely elastomeric Pneu-Nets:[1] i) These actuators can both fold and unfold; that is, they have positive curvature upon pressurization, and negative curvature under vacuum. ii) The texture at their gripping surface, from the fibers, gives them the ability to handle certain smooth materials[16] (e.g., glass) that would be difficult to manipulate with smooth surfaced grippers. iii) The bellows-like actuators do not require materials that are highly extensible (i.e., capable of hundreds of percent strains); they thus increase our range in materials choices for the design and fabrication of soft robots. (iv) They do not develop aneurysms over approximately two thousand cycles (we did not test to failure).

The material system we chose was polyaramid fibers (Kevlar™Pulp, FibreGlast Inc.; 15 wt%; Fig. S2) with lengths > 1.0 mm, and average diameters, d~100 μm, blended into an uncured silicone matrix (Ecoflex 0030™, Smooth-On Inc.; 85 wt%). Though cellulose would have also been a good choice, the polyaramid fiber was readily available in a size range that blended well (via mixing with an impeller blade for 15 minutes; see Supplemental Information,
SI, for more detail) with the uncured silicone. After blending, the mixture was a paste, but one that we could still pour into the molds. We cured the mixture (60 °C for 30 minutes), and then bonded the resulting bellows to an inextensible flat composed of the same material (see SI for more details). These blended composites are not as puncture resistant as woven Kevlar™, or as extensible as Ecoflex. As actuators, they have, instead, intermediate properties—resistance to puncture from many mundane hazards (e.g., broken glass, thorns on plants, and sharp rocks), and extensibility sufficient to achieve significant ranges of motion.

**Mechanical Properties**

Materials with large elastic moduli ($G'$) are difficult to puncture (e.g., diamond has an elastic stiffness modulus of ~124 GPa)\(^{[17]}\) and the composite material we used has four times the elastic modulus ($G'_{\text{comp}}\sim400$ kPa; Fig. S1) of the Ecoflex 0030™ ($G'_{\text{Eco}}\sim100$ kPa; Fig. S1) used in our prior publications.\(^{[1,4]}\) Also, the inclusion of fibers in rubber composites increases the surface area created during crack formation,\(^{[18]}\) and thus energy required to propagate the crack. In combination, these properties—high elastic modulus, large strain to failure (~200%; Fig. S1), and disruption of crack propagation—make the composite material actuators less sensitive to puncture than purely elastomeric ones. To compare the puncture resistance of the composite material and the pure elastomer, we pressed a steel cylinder (5.0 mm diameter) into sheets (5.0 mm thick) of the fiber-Ecoflex 00-30 composite, as well as pure Ecoflex 00-30 and found that it required ~2 MPa of pressure to puncture the fiber composite, and ~0.75 MPa to puncture the unreinforced elastomer (Fig. S3).
Actuator & Gripper Design

The pleats increase the volume of material that can participate in actuation; although each section of the composite expands to a smaller degree than the unreinforced elastomer, the folds increase the number of elements that actuate, and allow large amplitudes of motion (Fig. 1). To achieve the greatest possible range of motion, we maximized the number of “folds” per actuator, within the limits set by the resolution of our 3D printed molds: ~125 microns; Dimension Elite, Stratasys, Inc.) and used this design for all actuators in this paper (Fig. 1a-c). The final pleating design we used (Fig. 1c,d) allowed us to achieve similar radii of curvature with the fiber-reinforced Pneu-Nets using both positive and negative (with respect to atmospheric) pressure (Fig. 1d-f; the acute angles at the tips of the actuator allowed it to collapse into a semicircle under vacuum).

To demonstrate the utility of the pleated Pneu-Net design, we oriented three of the actuators at 120° to one another (Fig. 2a), and glued them together to build a gripper that is capable of motion in two directions (SI provides fabrication details). We used this gripper to pick up a wine glass (Fig. 2a-d) using positive pressure (~15 psi; ~100 kPa): the convex curvature on the interior of the gripper surface conformed to the convex exterior of the smooth glass. We also used the manipulator with negative pressure to pick up the wine glass by contact with its interior surface (Fig. 2e-h).

Self Sealing
In cases where a sharp object punctured an actuator, a soft seal formed spontaneously around the hole and this seal maintained the functionality of the Pneu-Net (Fig. 3a-c). Even when the object was removed, the actuator continued to function (Fig. 3c-d). The fibers probably prevent crack propagation\cite{18} from expanding the damage due to the piercing object, and thus limit the extent of the crack to that of the piercing object itself.

We believe there are three primary mechanisms that cause the self-sealing phenomenon: (i) When a crack is created by an object, it pushes against and strains the bulk elastomer. Due to silicone’s high resilience (Fig. S1), when the object is removed the silicone returns to its original shape and presses the fresh crack surfaces against themselves—thus sealing the hole. (ii) The crack surfaces are deformable and conform to one another. Chaudhury and Whitesides\cite{19} measured the self-adhesion of PDMS (Sylgard 170; Dow Corning, Inc.), and found a work of adhesion, $W$, to be $\sim 45 \text{ erg/cm}^2 \ (4.5 \mu\text{J/cm}^2)$ for that particular silicone elastomer; using that value of $W$, we calculate an adhesive energy of $\sim 9 \text{ erg} \ (0.9 \mu\text{J})$ for the hole created from the needle in Fig. 3 (see SI for more detail). (iii) While actuated, the internally pressurized bladder pushes the self-adhered crack inward and applies compressive force to seal the edges of the hole.

**Conclusion**

By using a stretchable composite of fibers and elastomer in a quasi-bellows geometry, and using soft lithography, we fabricated pleated, pneumatic actuators. These actuators could move in two directions using a single pneumatic control input, and were both soft and resistant to puncture; If punctured, they sealed around the piercing object, and also sealed the hole left
behind when the object was removed. We used these actuators to fabricate a gripper that was delicate enough to pick up a wine glass from either its inner or exterior surfaces.

The practicality of soft robots as assistants to humans in search and rescue, as aids in rehabilitation, assisted living, or other tasks that require they be insensitive to sharp objects such as glass, barbed wire, thorns, and sharp rubble. These pleated actuators—composites that are both puncture resistant and flexible—are potential components in such systems. The addition of fibers to the elastomeric matrix also prevented failure via the occurrence of aneurysms over the thousands of cycles we tested; this feature, combined with their reduced sensitivity to puncture, will make them useful choices for soft machines in areas that demand reliability in hazardous environments.

Finally, the bellows design we chose is a platform for using fibers and less extensible materials in soft actuators. For example, the use of carbon fiber would add electrical conductivity, and potentially even higher strength-to-weight ratios than the current choice of materials. Glass fibers \((G'_{SiO2} \sim 70 \, GPa)^{[20]}\) may improve their resistance to slashes, abrasion, and tearing (modes of failure we did not explore). In addition, this design allows us to use less extensible rubbers, such as the styrene-butadiene (SBR) formulations in tire rubber;\(^{[21]}\) by using these high elastic modulus rubbers \((G'_{SBR} \sim 80 \, MPa)^{[22]}\) it should be possible to actuate pneumatically to higher pressures, and thus to exert greater forces (semi-truck tires are routinely pressurized to >100 PSI; 690 kPa) than is possible in silicone-based soft machines.
Acknowledgements

This work was supported by DARPA under award number W911NF-11-1-0094. Robert Shepherd, Adam Stokes, and George Whitesides conceived of and performed the experiments and wrote the text. Rui Nunes performed the puncture tests and microscopy.

References

Supplemental Information

Soft Machines That are Resistant to Puncture, and That Self Seal

Robert F. Shepherd¹, Adam A. Stokes¹, Rui M.D. Nunes¹, George M. Whitesides¹,²*

¹ Department of Chemistry and Chemical Biology, Harvard University,
12 Oxford Street, Cambridge, Massachusetts 02138

² Wyss Institute for Biologically Inspired Engineering
60 Oxford Street, Cambridge, Massachusetts 02138

*Corresponding author, email: gwhitesides@gmwgroup.harvard.edu
Actuator & Gripper fabrication

We first blended the fibers (5 grams, ~50 ml dry volume) into Ecoflex 0030 (30 grams). The result of blending was a spongy paste that we then pressed between the two part mold (printed using a 3D printer; Dimension Elite) sketched in Fig. 1a. After 15 minutes in an oven at 70\(^\circ\)C, we pulled the mold apart and removed the pleated actuator (active layer; Fig. 1b) and sealed it to a flat sheet (strain limiting layer; Fig 1c) of the fibers impregnated with uncured silicone. After another heat treatment in the oven (15 minutes at 70\(^\circ\)C), we inserted the pneumatic tether (a silicone tube). To fabricate the gripper, we arrayed three of these actuators 120\(^\circ\) to one another and glued them together using silicone elastomer (Sylgard 184; Dow Corning, Inc.) in a mold cut to hold each of the three actuators.

Calculating the adhesive energy for a crack created by a puncture from a 14 gauge needle

The #14 needle (diameter = 2.1 mm) that punctured the actuator has a circumference of 0.66 cm and it penetrates a thickness of 0.5 cm (the membrane thickness). The surface area created (0.33 cm\(^2\)) and the work of adhesion, \(W\approx45\text{ erg/cm}^2\), of a typical silicone[1] causes a self-adhesive energy of 9 erg (0.9 \(\mu\)J).

Self-sealing gripper using elastomeric Pneu-Nets and hydraulic actuation

We also demonstrated a self-sealing gripper using elastomeric actuators (Fig. S4). This system works by using hydraulic actuation, where the liquid medium (in our case a commercially available tire sealant; Slime\textsuperscript{TM}) is filled with high aspect ratio fibers. When a puncture occurs in the in the Pneu-Net, while actuated, the pressure difference from the inside of the Pneu-Net to the outside (\(\Delta P = +\)) causes the hydraulic fluid to squeeze through the hole. The fibers jam as they squeeze through this puncture, sealing it (Fig. S5).
References

**Figure 1.** (a) Two part mold for soft lithography of a pleated, bellows-like actuator. Ecoflex™/Kevlar™ (composite, yellow) is pressed into the mold and (b) replicated, then pressed against and bonded to a composite flat. The arrows indicate the direction in which the (b) mold or (c) active layer is applied. (d) Fabricated composite bellows actuator with internal pneumatic network at ambient pressure (ΔP = 0), (e) positive pressure (~15psi; ΔP = +), and (f) negative pressure (house vacuum; ΔP = -). Bellows actuators with (g,h) ~0.5 pleats cm⁻¹ and (i,j) ~1.3 pleats cm⁻¹. Scalebars are 2 cm.
Figure 2. A gripper picks up a wine glass by gripping the (a-d) exterior surface or (e-h) interior surface.
Figure 3. (a) A composite material, bellows-like actuator that (b) bends when pressurized. (c) The material seals around a puncture and the actuator continues to function, (d) even when the source of puncture (a 14 gauge needle) is removed. Scalebar is 2 cm.
**Figure S1.** Stress vs. strain curves for a Kevlar/Ecoflex composite (black) and Ecoflex (gray) strip. Both materials were unloaded at 100% strain for one cycle to measure their resilience at that strain—there is no visible hysteresis in the graph. The loading was then reapplied until failure.
Figure S2. Optical microscopy images of (a) polyaramid fiber bundle and (b) individual fiber. Scalebar is 100 microns.
Figure S3. A 5 mm cylinder is pressed onto 5 mm thick sheets of (a-c) Ecoflex 0030 silicone elastomer and (d-f) a composite of polyaramid fibers and silicone elastomer until they are punctured. (g) The pressure applied to the sheets as the cylinder punctures the samples is shown for ecoflex (gray) and the fiber-silicone composite (black).
Figure S4. a) Schematic diagram of soft gripper assembly. b) Top face view of a gripper with a channel filled with slime™. c) Profile scheme of a gripper arm.
Figure S5. Photos of the hydraulic actuation of an elastomeric gripper and its puncture and self-sealing. a) non-actuated gripper, b) actuated gripper, c) puncturing the inflated side of the gripper with 3.76 mm diameter nail, d) punctured gripper maintains actuation, e) gripper still functions after the nail is removed.