"Elastomeric Origami: Programmable Paper-Elastomer Composites as Pneumatic Actuators"

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Composite Materials to Fabricate Soft Pneumatic Actuators. Flexible but nonstretchable materials can be embedded in Ecoflex can be used to fabricate soft pneumatic actuators as described in this work. **Figure S1**, **S2**, and **S3** show actuators fabricated with these kind of materials. Flexible materials with a bad adhesion to Ecoflex (such us polymer films) can be embedded into the elastomer by punching holes along their surface (Fig. 4E-F). All materials were purchased from McMaster–Carr Inc. (Chicago, IL). We embedded the materials in Ecoflex by pouring the prepolymer on top, degassing to remove air bubbles in a desiccator at 36 Torr for 3 min, and curing at 60 °C for 15 min. To connect the pneumatic network of the soft actuators with the external gas source we used polyethylene tubing (Intramedic, Sparks MD) with an outer diameter of 1.57 mm. This tubing was easily introduced into the soft actuator using a regular hypodermic needle (16G, 3.81 cm long) as the connector (process depicted in **Fig. S5**).

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Figure S1. Soft actuators that embed different flexible but nonstretchable materials that make them bend upon pressurization. a) Tulle. b) Cooper mesh. c) Cotton cloth. d) Nylon mesh.e) PET film. f) Polyester tape.



Figure S2. Soft actuator that embeds a thread parallel to the pneumatic channel. a) Schematic representation of the fabrication process. Both ends of this actuator are glued with Ecoflex to seal the pneumatic channel. b) Resting state (P_{atm}) of such a device. Inset in b) correspond to the optical image of the lateral cross section of the device. c) Actuated state (P_1 =75 mbar).



Figure S3. Soft actuator that embeds a thread rolled around the pneumatic channel. a) Schematic representation of the fabrication process. Both ends of this actuator are glued with Ecoflex to seal the pneumatic channel. b) Resting state (P_{atm}) of such a device. Inset in b) correspond to the optical image of the thread embedded in the actuator. c) Actuated state (P_1 =125 mbar).



Figure S4. Procedure followed to insert the tubing into a soft paper-Ecoflex actuator.a) 1.65 mm thick cannula. b) Polyethylene tubing with an outer diameter of 1.57 mm.c) Ecoflex-paper slab simulating the wall of the actuator that is going to be connected to the gas source by the tubing. The insertion of the tubing requires to perforate the wall of the actuator with the cannula, to both introduce the tubing through and to remove the cannula.



Figure S5. Connection to the external gas source. a) Components used to pressurize the pneumatic actuators with a 60 ml syringe (top). Final connection (bottom) . b) Elements used to pressurize the actuators with a compressed gas bottle (top). Elements after being connected (bottom).

Fabrication of Bellows Structures. Figures S6 and **S7** show the design and mechanical response of a bellows structure with an internal Ecoflex strip that links the top and the bottom caps of the actuator. The elastomeric strip is stretched when the actuator extends upon pressurization. When the internal pressure decreases, the elastic restoring force of the Ecoflex strip causes the device to recover its original shape without significant hysteresis.



Figure S6. Folding scheme to fabricate origami actuators. a) Folding marks for a rectangular single piece of paper. b) The folded piece of paper is first folded along the three vertical axis shown in a). The flap closes the structure by overlapping. c) Paper origami structure obtained by folding the paper along the creases defined in a). d) top view of the paper structure showed in c).



Figure S7. Origami actuator with the hysteresis compensated by the elastic recovery of an Ecoflex strip that joins the top and the bottom of the actuator. a) Top view of the origami structure without the top cap. The Ecoflex strip is glued with more Ecoflex to the bottom cap of the actuator. b) Folded actuator in the resting state (atmospheric pressure inside the pneumatic chamber). The inset shows the relaxed piece of Ecoflex linking the top and the bottom of the actuator. c) Extended actuator when the pressure in the pneumatic chamber is 175 mbar. The inset shows the stretched piece of Ecoflex that is housed in the actuator. d) Pressure dependence of the extension of the actuator relative to its length at rest. In this case, the Ecoflex strip housed into the actuator helps the actuator to fold up when the pressure decreases minimizing the hysteresis.

Cost. We have not yet considered the issues that would arise in manufacturing. Excluding labor and capital expenses, however, the estimated cost for making any of the actuators described in this communication is less than \$10: i) The estimated cost of the molded material is less than \$1.5 (<15g at \$0.10/g for silicone-based materials). ii) The estimated cost for printing the reusable 3D mold used to fabricate some of the actuator described is less than \$7 (22 g at \$0.30/g for the 3D printed material). iii) The estimated cost of the paper used to make the actuators is less than \$0.01 (<100 cm² at \$0.44/m²).

Scanning electron imaging. Scanning electron microscope (SEM) image (Fig. 5B inset) of the paper-elastomer actuator was acquired with a Zeiss Supra55 VP FESEM at 2 kV at a working distance of 6 mm. Before SEM imaging, the sample was placed on a silicon wafer and sputter coated with Pt/Pd at 60 mA for 15–45 s.

Luminous intensity measurements. We used a high-speed silicon photodetector (Thorlabs DET 10A) to measure the light emission from the soft actuator shown in Figure 10. The output signal from the photodetector (in the 0-10V range) was registered by a digital oscilloscope (Tektronix TDS 2024B). Both soft actuator and photodiode were isolated from any external light and kept at a separation distance of 1 m during the measurements.