

*Supplemental Information*

*for*

**Elastomeric Tiles for the Fabrication of Inflatable Structures**

Stephen A. Morin<sup>1</sup>, Sen Wai Kwok<sup>1</sup>, Joshua Lessing<sup>1</sup>, Jason Ting<sup>1</sup>, Robert F. Shepherd<sup>1</sup>, Adam

A. Stokes<sup>1</sup>, and George M. Whitesides<sup>\*1,2,3</sup>

*Department of Chemistry and Chemical Biology, Harvard University*

*12 Oxford Street, Cambridge, MA 02138*

\* Author to whom correspondence should be addressed

<sup>1</sup>Department of Chemistry and Chemical Biology  
Harvard University

12 Oxford Street, Cambridge, MA 02138, USA

Email: gwhitesides@gmwhgroup.harvard.edu

<sup>2</sup>Kavli Institute for Bionano Science & Technology  
Harvard University

29 Oxford Street, Cambridge, MA 02138, USA

<sup>3</sup>Wyss Institute for Biologically Inspired Engineering  
Harvard University

60 Oxford Street, Cambridge, MA 02138, USA

## Materials and Methods

*Materials:* We used all materials as received unless otherwise noted. We purchased polydimethylsiloxane (PDMS, Sylgard<sup>®</sup> 184, Dow Corning Corporation<sup>®</sup>) and Ecoflex<sup>®</sup> (Shore hardness 0030, Smooth-on Inc.) two-part silicone elastomer kits from Ellsworth Adhesives and Reynolds Advanced Materials respectively. We purchased acrylonitrile butadiene styrene (ABS) cartridges for use with a Dimension Elite (manufactured by Stratasys Ltd.) three-dimensional (3D) printer from Stratasys Ltd. We purchased polyethylene tubing (I.D. 0.045", O.D. 0.062", PE 160, BD Intramedic<sup>™</sup>) and silicone tubing (I.D. 0.030", O.D. 0.065", VWR International) from VWR International LLC. We purchased barbed male and barbed female Luer lock fittings (for tube I.D. 0.06", nylon) from McMaster-Carr<sup>®</sup>. We purchased an assorted kit of light emitting diodes (LEDs, through hole mount, round, 5mm, terminal 1.75", Rohm Semiconductor, #511-8001-KIT) and wire (AWG 40, silver-coated copper, polytetrafluoroethylene insulated) from DigiKey<sup>®</sup>. We purchased steel wool (grade "0000," average fiber width ~ 25  $\mu$ m) from Steel Wool International. We purchased silver conductive adhesive paste (478SS, #12685-15, Electron Microscopy Sciences) directly from the manufacturer. We purchased the dyes hexamethylpararosaniline chloride (Crystal Violet, CAS# 548-62-9, ACS Reagent, >90% dye content), 2,4-dihydroxyazobenzene (Sudan Orange G, CAS# 2051-85-6, 85% dye content), and Thioflavin T (CAS# 2390-54-7, 65-75% dye content) from Sigma-Aldrich<sup>®</sup>.

*Design of the Masters for Tile Fabrication:* We demonstrated structures that were fabricated using single-taper or double-taper dovetail joints designs. For the fabrication of cubes with single-taper dovetail joints we used a set of tiles consisting of three unique designs (Figure 1C main text)—tiles on opposing faces of the cube were identical to each other but matting tiles were not. We required a set of three masters for the fabrication of these tiles (Figure S1).

Though the faces of a cube are geometrically indistinguishable, we arbitrarily refer to the single-taper masters (and the tiles they are used to fabricate) as “top” and “bottom” (Figure S1A) and “sides” (Figure S1A, B). For the fabrication of cubes with double-taper dovetail joints we used a set of tiles consisting of two unique designs. Again, tiles on opposing faces of the cube were identical, but in this design only two tiles mate with unique tiles. We required a set of two masters for the fabrication of these tiles (Figure S2). The double-taper masters (and the tiles they are used to fabricate) are referred to as “top” and “bottom” (Figure S2A) and “sides” (Figure S2B). The mating requirements of dovetail joints (pins only fit with tails) in the fabrication of a cube necessitate at least two unique tiles for these structures and the double-taper design is at this minimum.

*Fabrication of the Masters for Molding Elastomeric Tiles:* We fabricated the masters for the elastomeric tiles from ABS thermoplastic using a three dimensional (3D) printer (Dimension Elite manufactured by Stratasys, Ltd.). We saved the CAD designs for the masters in the standard tessellation language (.stl) file format and loaded them into the printer for production using a specialized software package provided with the printer (CatalystEX from Stratasys, Ltd.). The masters were used as received from the printer without further modification.

*Fabrication of Elastomeric Tiles with Homogeneous Materials Composition:* We fabricated the tiles for the cubes from polydimethylsiloxane (PDMS), Ecoflex<sup>®</sup> (0030), or combinations of both materials. To fabricate elastomeric tiles with homogeneous materials composition we first prepared liquid PDMS or Ecoflex<sup>®</sup> pre-polymer following the instructions provided by the manufacturer (10:1 ratio of base to curing agent for PDMS and a 1:1 ratio of part A to part B for Ecoflex<sup>®</sup>). We then filled each master with an appropriate amount of liquid pre-polymer (~4 ml) so that the level of the polymer was even with the top of the mold. We transferred the pre-

polymer-filled masters to an oven for thermal curing at 60° C. We leveled the shelves of the oven prior to transfer to insure the tiles were of even thickness. After 40 minutes or 15 minutes for PDMS or Ecoflex<sup>®</sup> respectively, we removed the cured tiles from the oven and released them from the master by carefully prying them out with a pair of blunt-nosed tweezers. If necessary, we used a pair of fine-tipped microscopy scissors to trim the edges of the tiles clean of any imperfections (e.g., from slightly overfilling the master).

*Fabrication of Elastomeric Tiles with Heterogeneous Materials Composition:* We fabricated heterogeneous tiles by first fabricating homogeneous tiles as described above, but we did not remove the tile from the mold after curing. Instead, we used a razor blade to cut the desired pattern into the tile (e.g., a circle) while it was still inside the mold. This approach was much easier than removing the tiles and cutting them because the tiles are difficult to keep steady during cutting because they are thin and easily deformed. Once the desired pattern was cut into a tile, the unwanted portion of the tile was carefully removed with a pair of tweezers. Next, we filled the voids in the tiles using either PDMS or Ecoflex<sup>®</sup> prepolymer and transferred them (while still in the master) to the oven for thermal curing. After 40 minutes or 15 minutes of curing for PDMS or Ecoflex<sup>®</sup> respectively, we removed the cured heterogeneous tiles from the oven and released them from the master by carefully prying them out with a pair of blunt-nosed tweezers. If necessary, we used a pair of fine-tipped microscopy scissors to trim the edges of the tiles clean of any imperfections (e.g., from slightly overfilling the master).

*Fabrication of Colored Elastomeric Tiles:* We fabricated colored tiles following the molding procedure described above, except we used silicone pre-polymers that we colored with visible wavelength dyes. We made the colored pre-polymers by first dissolving the appropriate amount of the chosen dye (crystal violet for purple tiles, Sudan Orange G for orange tiles, and thioflavin

T for amber tiles) in dichloromethane to achieve an approximate concentration of 3 mg/mL (concentration of the solution of thioflavin T is 0.3 mg/mL due to its lower solubility in dichloromethane). We added this colored solution to silicone pre-polymers that had been prepared according to the manufacturer's instruction (described above) to achieve a ratio of approximately 1 mL of dye solution per 22 g of pre-polymer and mixed them thoroughly using an overhead stirrer equipped with a three-paddle propeller. We degassed this mixture in a vacuum desiccator at 65 kPa to remove most of the dichloromethane. The duration of degassing was 5 minutes for Ecoflex and 20 minutes for PDMS pre-polymer. We then used the resulting colored pre-polymer to fabricate colored tiles.

*Fabrication of the Hollow Cube Structures:* We fabricated a set of six complementary elastomeric tiles using the set of masters for the single-taper or the double-taper dovetail joint designs (the tiles for these different designs were not interchangeable; see the above section on the design of the masters for details). For the single-taper dovetail design this set included: two tiles fabricated from single-taper master A (Figure S1A) that form the “top” and “bottom” (Figure 1C, D; orange tiles) of the cube, two tiles fabricated from single-taper master SB (Figure S1B) that form two of the “sides” (Figure 1C, D; purple tiles) of the cube, and two tiles fabricated from single-taper master SC (Figure S1C) that form the remaining two “sides” (Figure 1C, D; yellow tiles) of the cube. For the double-taper dovetail design this set included: two tiles fabricated from double-taper master A (Figure S2A) that form the “top” and “bottom” of the cube, and four tiles fabricated from double-taper master B (Figure S2B) that form the four “sides” of the cube.

Before assembling the tiles into cubes, we identified an optimal location for the pneumatic inlet/outlet tube in the structure to be assembled. This location was ideally a place on the final

structure that was not expected to expand significantly during inflation because this choice minimized the potential for leaks at the inlet/outlet when the structure was inflated. For most structures the location chosen was toward the corner of one of the PDMS tiles. When there were no PDMS tiles or when the tiles that included PDMS had this material toward the center, we chose a location close to one of the corners. We intentionally did not choose the tile that would become the “bottom” face. Once the appropriate location was identified we used a 1.0 mm biopsy knife to cut a small hole in the tile. We did not insert a tube through the hole at this stage because it would complicate assembly of the tiles.

Regardless of the dovetail design, we assembled the tiles in the same order and used the same methods to glue them together. Fabrication progressed as shown in Figure 1D of the main text:

First, we coated the mating edges of the four tiles that interlock together to form the sides of the cube (Figure 1D; steps i-iii) with liquid pre-polymer (for gluing PDMS tiles to PDMS tiles we used PDMS pre-polymer; for gluing PDMS tiles to Ecoflex<sup>®</sup> tiles we used PDMS pre-polymer; for gluing Ecoflex<sup>®</sup> tiles to Ecoflex<sup>®</sup> tiles we used Ecoflex<sup>®</sup> pre-polymer) using a syringe tipped with an 18 gauge needle. Note that at this point we did not coat the edges of the tiles that interlock with the top or bottom tiles with liquid pre-polymer. We then transferred the assembled tiles that form the “sides” of the cube to an oven for thermal curing at 60° C. After 40 minutes or 15 minutes for PDMS or Ecoflex<sup>®</sup> pre-polymer glue respectively, we removed the cured assembly of tiles that form the sides of the cube from the oven and let it cool to room temperature. Next we coated the four edges of this assembly that mate together with the edges of the tile that forms the bottom of the cube as well as the four edges of the bottom tile itself (Figure 1D; steps iii-iv) with liquid pre-polymer (again using a syringe). We then transferred this five-tile assembly to an oven for thermal curing at 60° C. After 40 minutes or 15 minutes for PDMS

or Ecoflex<sup>®</sup> pre-polymer glue respectively, we removed the cured assembly of five tiles and let it cool to room temperature. Finally, we coated the remaining edges of this assembly of five tiles and the edges of the top tile with liquid pre-polymer and completed the cube assembly by locking the top tile in place (Figure 1D; step iv). The finished assembly of tiles was transferred to an oven for thermal curing at 60° C. After 40 minutes or 15 minutes for PDMS or Ecoflex<sup>®</sup> pre-polymer glue respectively, we removed the final cube structure from the oven and let it cool to room temperature. It was important to insure that the hole for the inlet/outlet tube was not blocked by liquid pre-polymer or otherwise covered during the final curing step because the hole (at this stage) functions as a vent for the release of air that expands inside the structure during heating. If this vent was not present the expanding air will escape through the joints compromising the formation of air-tight edges.

*Pneumatic Inflation/Deflation of the Structures:* We inserted a single polyethylene tube (I.D. 0.045", O.D. 0.062") through the previously cut inlet/outlet hole in order to inflate or deflate the structure using compressed air or vacuum respectively. We did not use glue to seal the tube, rather, because the whole was intentionally made smaller than the O.D. of the tube, the elastomeric tiles formed a compression seal around the tube.

We changed the shape of the cubes by inflating them, using compressed air, at pressures ranging from 7 to 140 kPa above atmospheric pressure, or by evacuating them, using house vacuum, to at pressure of approximately 65 kPa. We used both computer-controlled and manually operated solenoid valves to control gas flow in and out of the structures.

*Peg/recess Connector for the Assembly of Extended Structures:* We connected the inflatable cube structures using a press connector of a design similar to one we reported previously (ref. 26, main text). One half of the connector consists of an array of pegs that mate with a matching

array of recesses on the other half of the connector (Figure 5 main text and Figure S3). We fabricated the connectors from PDMS. We first prepared liquid PDMS pre-polymer (or colored pre-polymer as described above if colored connectors were needed) following the instructions provided by the manufacturer (10:1 ratio of base to curing agent). We then filled each master (Figure S4A and S4B) with an appropriate amount of liquid pre-polymer (~3 ml) so that the level of the polymer was even with the top of the mold. We transferred the pre-polymer-filled masters to an oven for thermal curing at 60° C. We leveled the shelves of the oven prior to transfer to insure the connectors were of even thickness. After 40 minutes, we removed the cured connectors from the oven and released them from the master by carefully prying them free with a pair of blunt-nosed tweezers.

We used the connectors in three ways: (i) We glued the pegs to one cube and the recesses to another cube enabling the direct connection of the structures (Figure 5D), (ii) We glued two recess parts back to back forming a single part with recesses on either side that we used to connect cubes with pegs (Figure 5A-C), or (iii) We glued together recess parts along their lateral edges to create recess “strips” to which cubes with pegs could be attached (Figure 5E). We glued pegs/recesses to the cubes or recess parts to one another (back to back or laterally) using liquid PDMS pre-polymer. The interface of the two parts were coated with a layer of pre-polymer and pressed together. We then transferred the assembly to the oven for thermal curing at 60° C for 40 minutes. We glued pegs/recesses to the cube structures at PDMS faces. We focused on demonstrations that highlight reconfigurability and the peg/recess interfaces of the connectors were operated without glue, however, for applications where permanence is required they can be glued together.



*Fabrication of Tiles with Conductive Patches:* We began by fabricating a conductive composite comprised of Ecoflex<sup>®</sup> and steel wool. First we rolled a pad of steel wool into a cylinder and then inserted it into a 15 ml, conical centrifuge tube leaving approximately 2.5 cm of empty space from the mouth of the tube to the steel wool. We then replaced the air pockets in between the fibers with Ecoflex<sup>®</sup> pre-polymer (prepared according to the manufacturers recommendations) by repeating the following sequence as many times as necessary: pre-polymer was poured into the empty space at the top of the tube and then the tube was degassed in a vacuum desiccator at 65 kPa for 2 minutes. After we had replaced all the air with pre-polymer we transferred the tube to an oven for thermal curing at 60° C for 30 minutes. We then removed the tube and cut it length-wise to free the composite. We sectioned this composite into circular discs that were ~1.5 cm in diameter (the diameter of the tube) and ~5 mm in thickness.

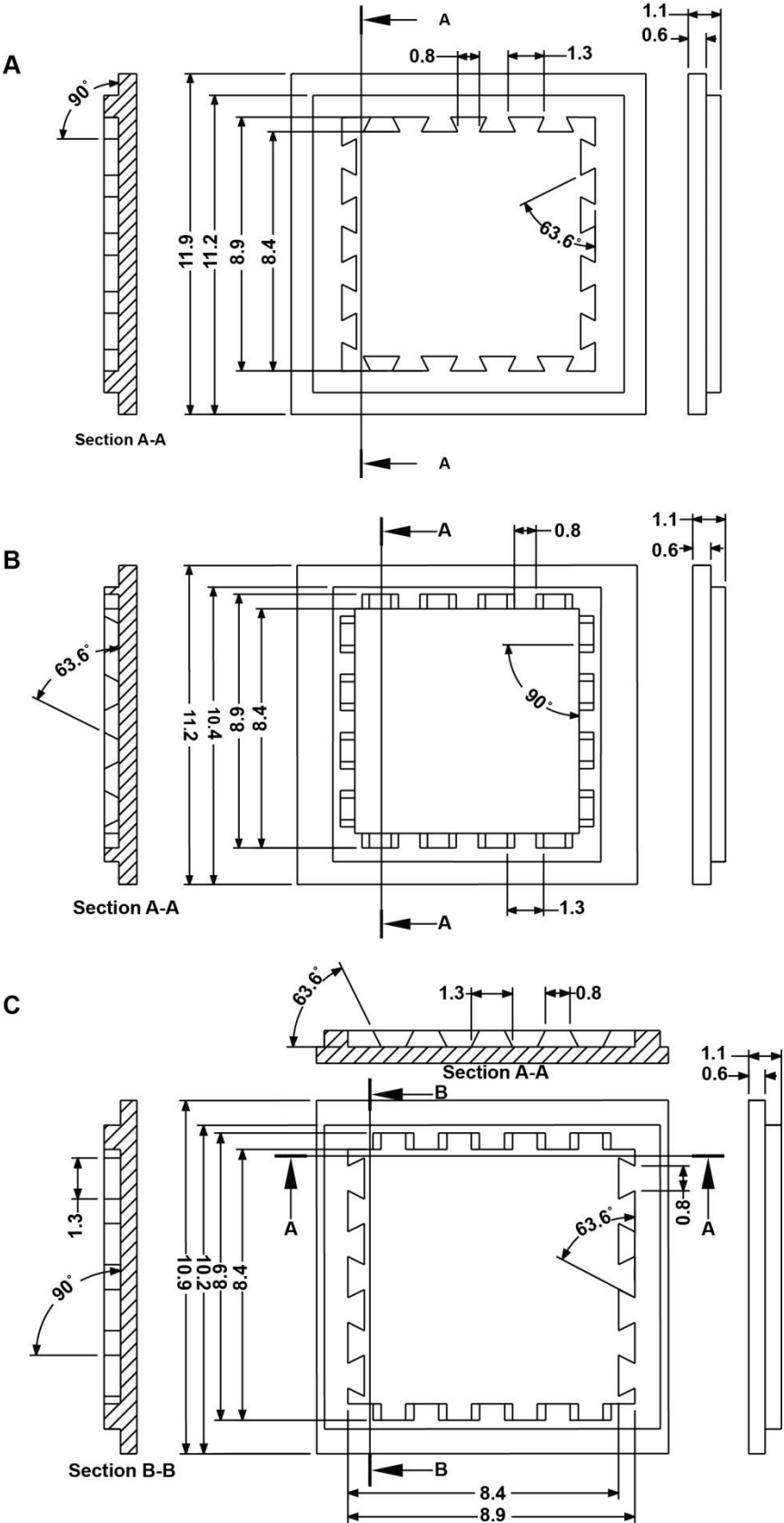
We fabricated tiles with conductive patches formed from these discs by positioning a disc in the center of the tile master and clamping it between two circular magnets (one on top of the disc and the other underneath the master). We then filled the master remaining volume of the master with Ecoflex<sup>®</sup> pre-polymer and moved it to an oven for thermal curing at 60° C. We removed cured tile from the oven after 15 minutes, removed the magnets, and carefully freed it from the master using a pair of blunt-nosed tweezers. If necessary, we used a pair of fine-tipped microscopy scissors to trim the edges of the tiles clean of any imperfections (e.g., from slightly overfilling the master).

*Fabrication of an Inflatable Cube with Embedded LED:* We first fabricated a complete set of six tiles with conductive patches in their centers (as described above) using either the single-taper or double-taper dovetail masters (Figure S1 and S2). Next, we soldered three 10 cm long wires (40 AWG) to each lead of a standard, through-hole-type, 5 mm, light emitting diode

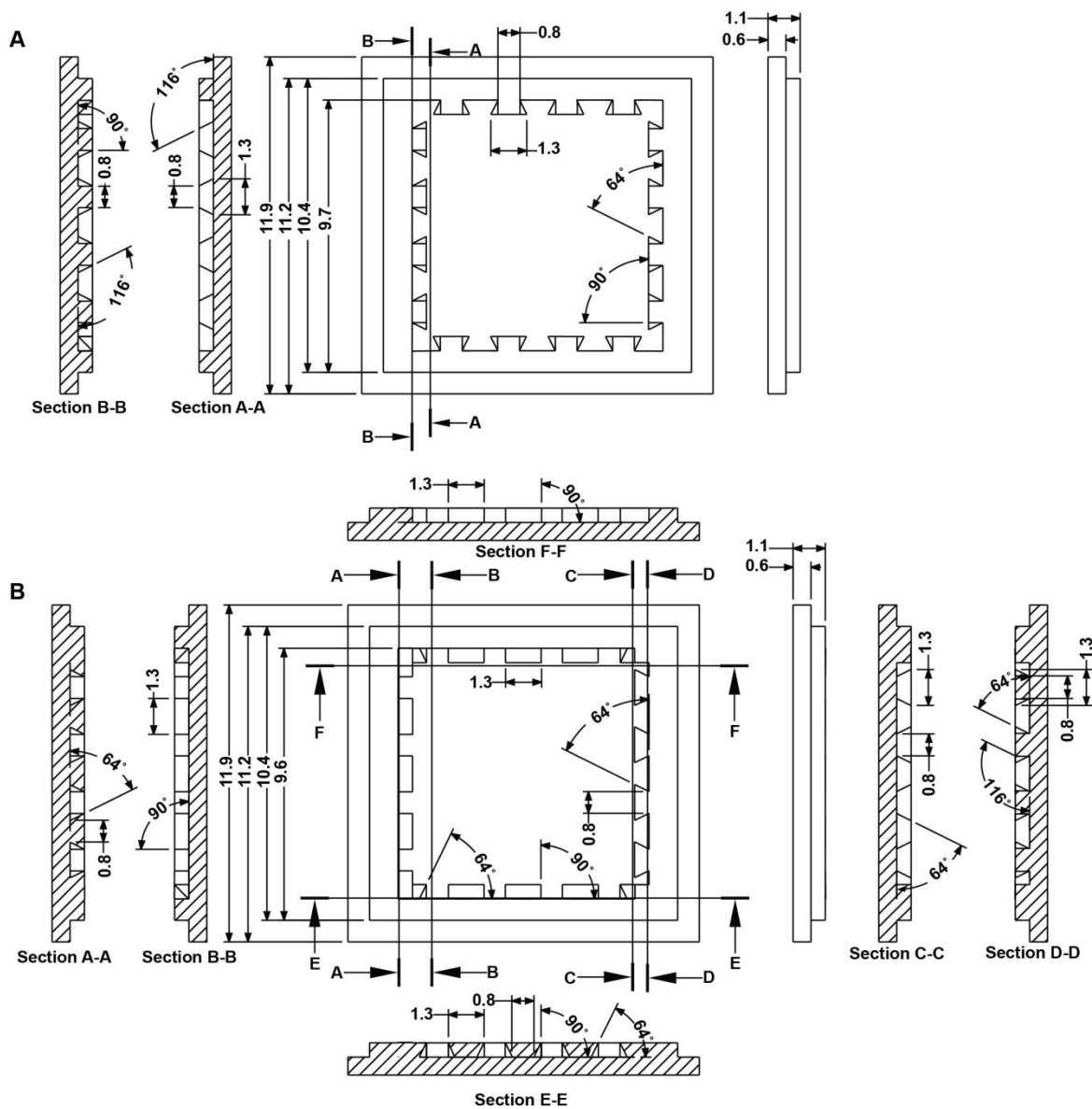
(LED). We then assembled a cube following the procedure above, except here we connected the LED across opposite faces of the structure by gluing the free ends of the wires to the interior side of each tile at the location of the conductive patch using silver conductive adhesive. We did this in the following way: (i) Cube assembly begins with gluing the “side” tiles together, so we started by attaching the four appropriate wires (two from the LED anode and two from the LED cathode connected to opposite faces) to the tiles that would make up the sides and letting the silver conductive adhesive cure at room temperature for 2 hours. We then glued the edges of the side tiles together and cured them as described previously. (ii) Next we attached one of the remaining wires to the “bottom” tile and allowed the silver conductive adhesive to cure at room temperature for 2 hours. We then glued the side assembly to the bottom tile as described previously. (iii) Finally we attached the last wire to the “top” tile and allowed the silver conductive adhesive to cure at room temperature for two hours. We then glued the top tile to the five-tile assembly (four sides and bottom) as described previously. We then tested the function of the internal LED by applying a forward bias to it through the conductive patches of the three sets of opposing faces of the cube using a 3 V power supply (current-limited to 10 mA) and a set of stainless steel probes.

**Figure S1.** Technical drawings of the three masters we designed, fabricated, and used to mold the tiles needed to assemble cubes with single-taper dovetail joints. (A) Master for molding the top and bottom tiles of the cubes. (B) Master for molding two of the side tiles of the cube and (C) the master for molding the other two side tiles of the cube. All dimensions are in centimeters.

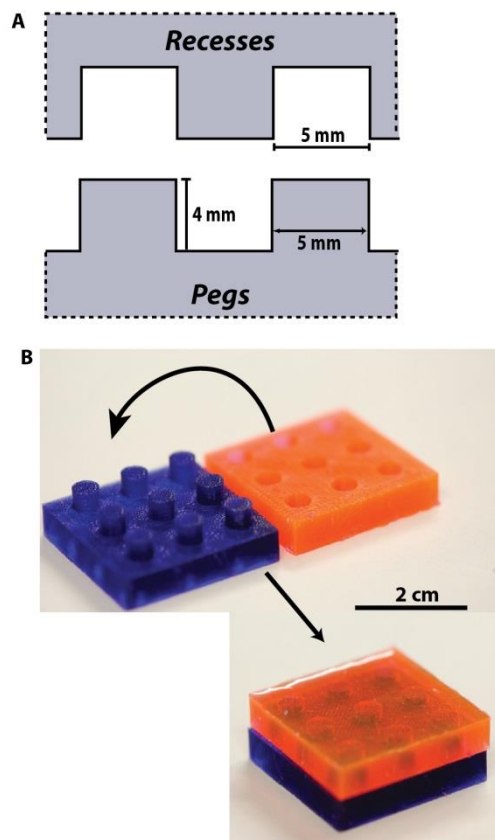
Figure S1.



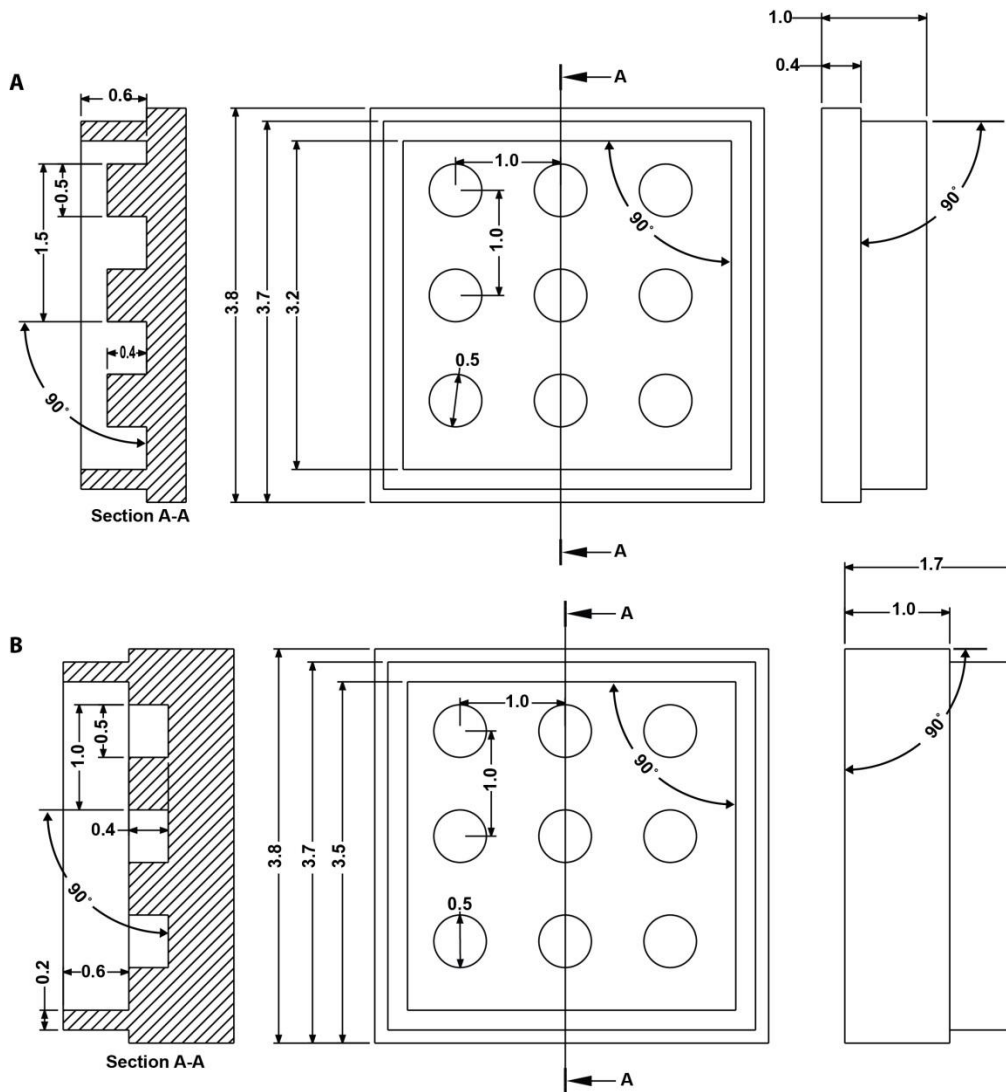
**Figure S2.** Technical drawings of the two masters we designed, fabricated, and used to mold the tiles needed to assemble cubes with double-taper dovetail joints. (A) Master for molding the top and bottom tiles of the cubes. (B) Master for molding two of the four side tiles of the cube. All dimensions are in centimeters.



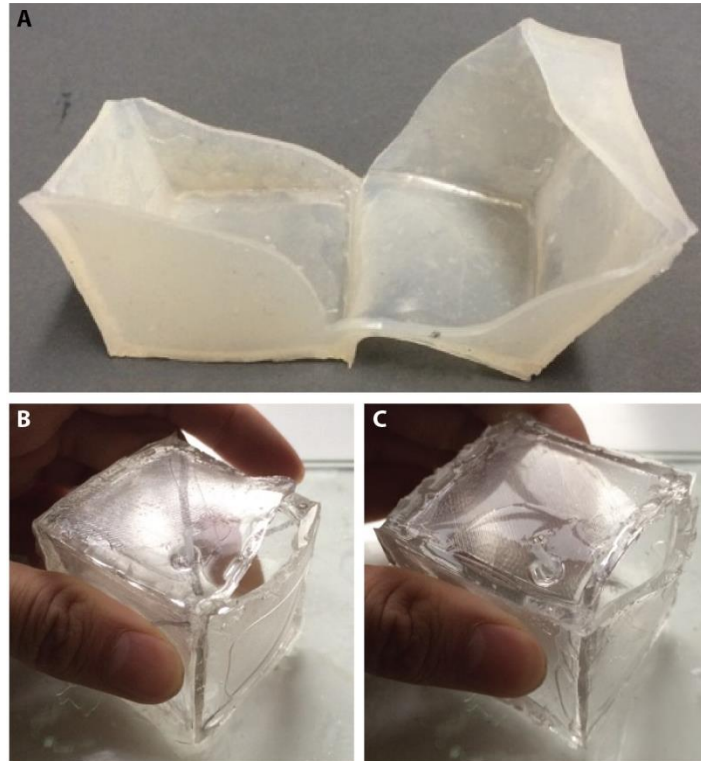
**Figure S3.** Peg/recess connector we used to assemble the inflatable cubes into extended structures. (A) Schematic illustration of the cross-section of the peg/recess connector. (B) Photographs of the connector before (top) and after we assembled it (bottom). To clearly illustrate the operation of the connector, we photographed both parts of the assembly (the violet pegs and the orange recesses) before we attached them to a cube.



**Figure S4.** Technical drawings of the two masters we designed, fabricated, and used to mold the complimentary peg and recess parts we connected the cube structures. (A) Master for molding the peg half of the connector. (B) Master for molding the recess half of the connector. All dimensions are in centimeters.



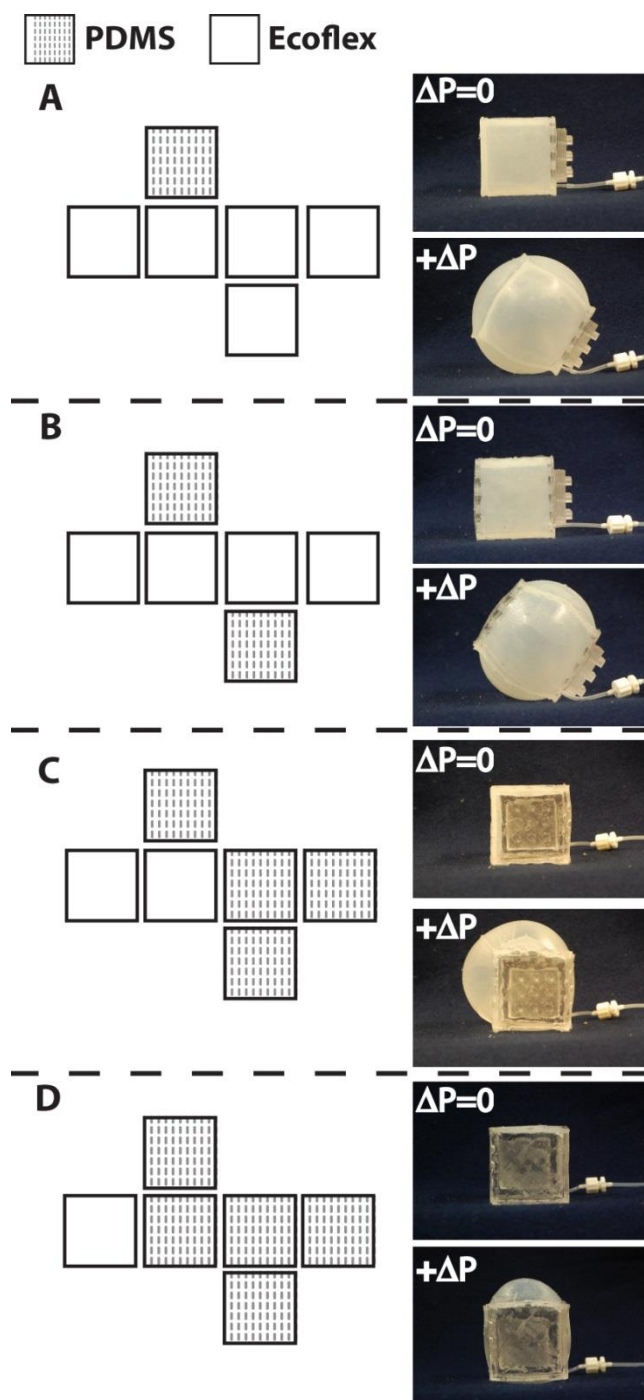
**Figure S5.** Failure of inflatable structures at elevated pressures. (A) Photograph showing a cube (assembled from all Ecoflex<sup>®</sup> tiles using single-taper joints; Figure 2A of the main text) after it exploded from over pressurization. Failure occurred when an internal pressure of  $\sim 120$  kPa ( $+\Delta P \sim 20$  kPa) was reached. The Ecoflex<sup>®</sup> material, not the soft joints, failed. (B) Photograph showing a cube (assembled from all PDMS tiles using single-taper joints) after it exploded from over pressurization. Failure occurred at the joint when an internal pressure of  $\sim 125$  kPa ( $+\Delta P \sim 25$  kPa) was reached. (C) Photograph showing a cube (assembled from all PDMS tiles using double-taper joints) after it exploded from over pressurization. Failure occurred at the joint when an internal pressure of  $\sim 140$  kPa ( $+\Delta P \sim 40$  kPa) was reached. For scale, the edge of each cube is 4.5 cm in length.



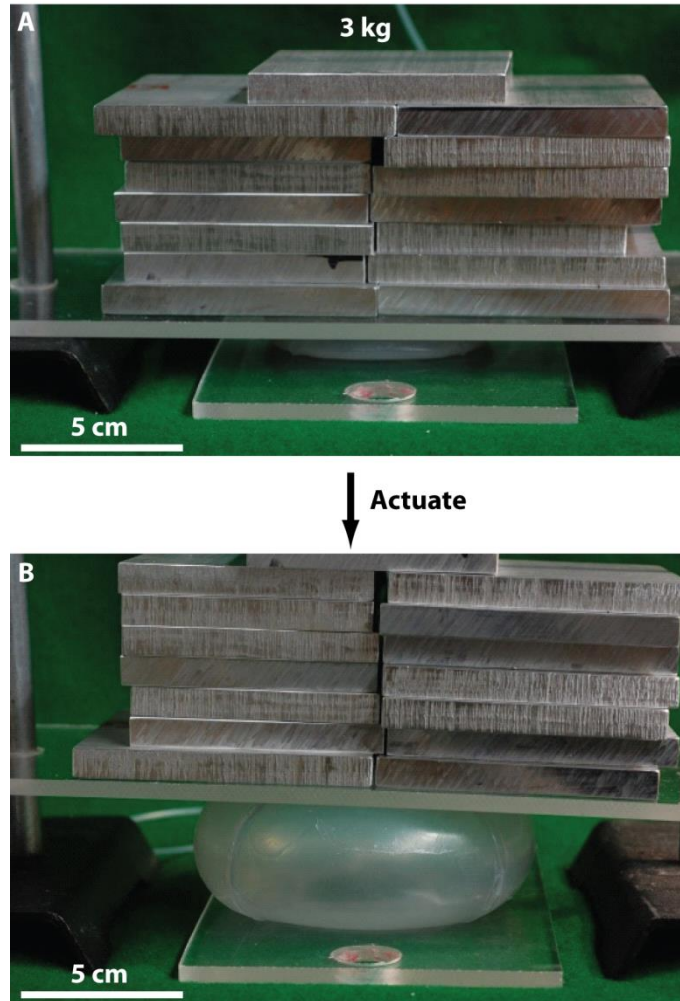


**Figure S6.** (A-D) Shape-changing structures we fabricated from tiles of varying extensibilities. Each panel includes a schematic that illustrates the material used for each tile (PDMS is hatched and Ecoflex<sup>®</sup> is white). We photographed the structures under isobarometric ( $\Delta P=0$ ) pressure (top panels) and the structures under positive ( $+\Delta P$ ) pressure (bottom panels) to illustrate shape change. We photographed the cubes oriented in the direction that illustrates change of shape most clearly. For scale, the non-deformed edges of the cubes are all 4.5 cm. We used a positive pressure of  $+\Delta P \sim 10$  kPa in these demonstrations.

Figure S6.

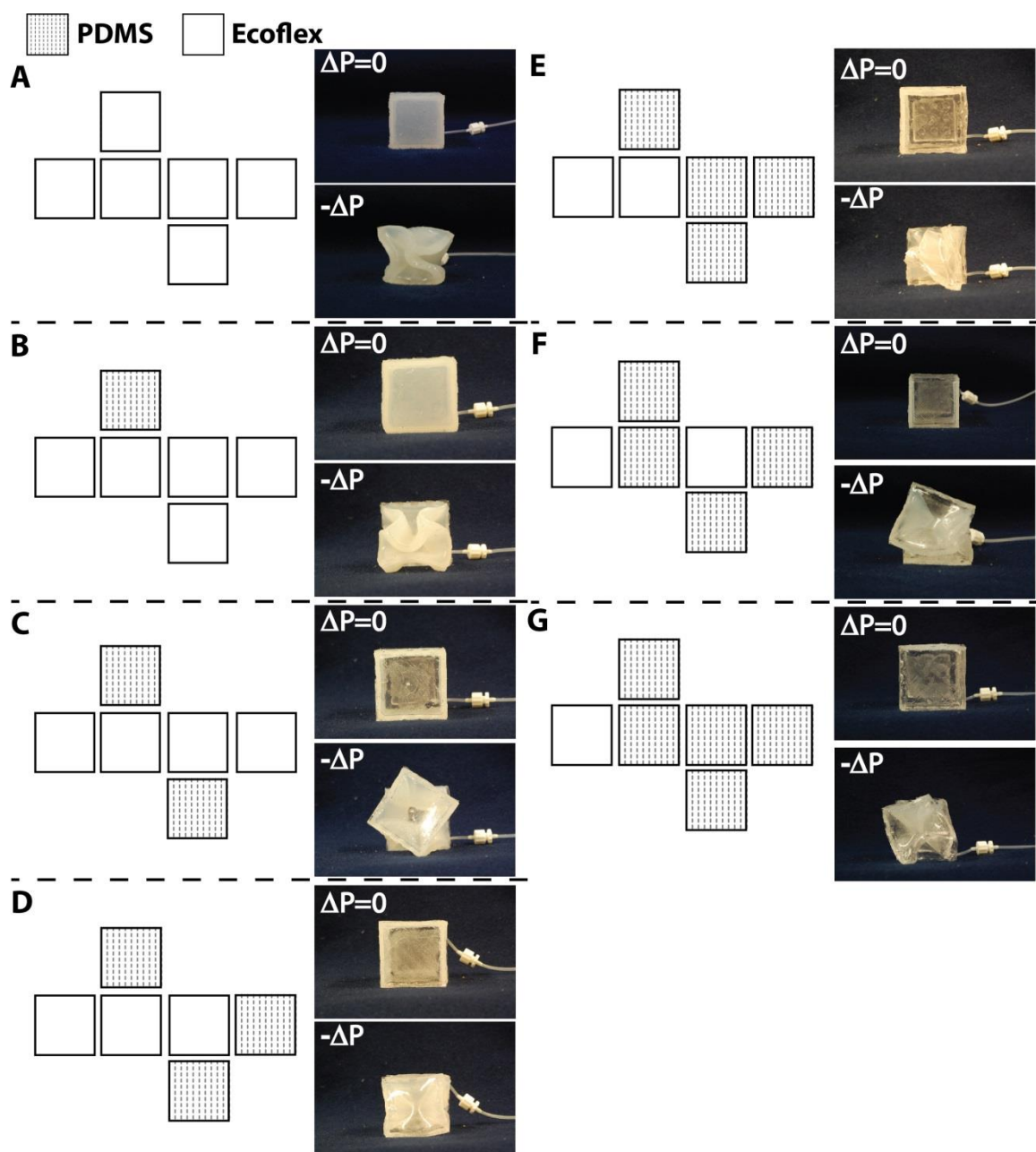


**Figure S7.** (A) We collapsed a cube structure under the weight of 3 kg. (B) We inflated the structure with a positive pressure of  $+\Delta P \sim 100$  kPa until the weight was lifted approximately 4 cm and then we closed the source valve (at this point we estimated the interior pressure of the device to be  $\sim 115$  kPa,  $+\Delta P \sim 15$  kPa relative to atmospheric pressure, based on the mass of the load and the surface area of the cube face). We could have used less pressure to inflate the cube and this approach would have lifted the weight at a slower rate. The cube weighed 30 g, a factor of 100 less than the weight lifted.



**Figure S8.** (A-G) Buckling of shape-changing structures. We made each tile from one type of silicone rubber; either PDMS or Ecoflex<sup>®</sup>, which has a lower elastic modulus and higher elastic limit. Schematics are included that illustrates the material composition and layout of the tiles in each structure (PDMS is hatched and Ecoflex<sup>®</sup> is white). We photographed the structures under isobarometric ( $\Delta P=0$ ) pressure (top photos) and the structures under negative ( $-\Delta P$ ) pressure (bottom photos) to illustrate shape change. We photographed the cubes oriented in the direction that illustrates change of shape most clearly. For scale, the non-deformed edges of the cubes are all 4.5 cm. We used a negative pressure of approximately  $-\Delta P \sim 35$  kPa to deflate the structures.

Figure S8.



## Supporting Videos

**Video S1.** A cube assembled from elastomeric tiles (all Ecoflex<sup>®</sup>, Figure 2A main text) expands into a sphere when ~10 kPa of compressed air is applied to the interior (real-time playback).

**Video S2.** A cube assembled from elastomeric tiles patterned with two different elastomers expands anisotropically when ~10 kPa of compressed air is applied to the interior (playback is in real time except deflation from 14 s to 17 s where playback is 4X real time).

**Video S3.** A cube assembled from elastomeric tiles patterned with two different elastomers illustrates controlled buckling when negative pressure (approximately –35 kPa) is applied to the interior (playback is in real time except deflation from 9 s to 12 s where playback is 4X real time and re-inflation, using atmospheric pressure, from 22 s to 25 s where playback is 8X real time).

**Video S4.** We collapsed and raised a tower constructed by stacking three cubes by sequentially deflating and reinflating the individual components of the structure (playback of collapse and re-inflation are real time; playback was sped up when the pneumatic lines are switched from positive to negative pressure at 12 s).

**Video S5.** We inflated a cube with conductive faces connected by an internal LED inducing contact of the cube with two charged surfaces (the top was a metal screen and the bottom was an aluminum sheet) that were held at a potential difference of 3V. This condition forward biased the LED and caused it to light up. We then rolled the cube between orientations that forward and reverse biased the LED causing it to turn on and off respectively (real-time playback).