



An integrated design and fabrication strategy for entirely soft, autonomous robots

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1 **An Integrated Design and Fabrication Strategy for Entirely Soft, Autonomous Robots**

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16
17 **Soft robots possess many attributes that are difficult, if not impossible, to realize with**
18 **conventional robots composed of rigid materials.^{1,2} Yet, despite recent advances, soft robots**
19 **still remain tethered to hard robotic control systems and power sources.³⁻¹² New strategies**
20 **for creating completely soft robots, including soft analogs of these crucial components, are**
21 **needed to realize their full potential. Here, we report the first untethered operation of a**
22 **robot comprised solely of soft materials. The robot is controlled with microfluidic logic¹³**
23 **that autonomously regulates the catalytic decomposition of an on-board monopropellant**
24 **fuel supply. Gas generated from fuel decomposition inflates fluidic networks downstream**
25 **of the reaction sites, resulting in actuation.¹⁴ The robot's body and microfluidic logic are**
26 **fabricated by molding and soft lithography, respectively, while the pneumatic actuator**
27 **networks, on-board fuel reservoirs and catalytic reaction chambers needed for movement**
28 **are patterned within the body via a multi-material, embedded 3D printing technique.^{15,16}**
29 **The relevant length scales of fluidic and elastomeric architectures required for function**
30 **spanned several orders of magnitude. Our integrated design and rapid fabrication**

31 **approach enables the programmable assembly of multiple materials within this**
32 **architecture, laying the foundation for completely soft, autonomous robots.**

33 Soft robotics is a nascent field that aims to provide safer, more robust robots that interact
34 with humans and adapt to natural environments better than their rigid counterparts. Unlike
35 conventional robots composed of rigid materials, soft robots based on hydrogels,^{17,18}
36 electroactive polymers,¹⁹ granular media,²⁰ and elastomers^{5,21} exhibit elastic moduli ranging from
37 100kPa to 1MPa,¹ are physically resilient,^{22,23} and have the ability to passively adapt to their
38 environment.^{1,2} Molded and laminated elastomers with embedded pneumatic networks are
39 widely used materials in soft robotics.^{1,21} Actuation of these elastomeric composites occurs when
40 interconnected channels comprising the pneumatic network are inflated with incompressible
41 fluids or gases supplied via tethered pressure sources.¹ Robotic end effectors with bioinspired⁷
42 and rapid²⁴ actuation, deployable crawlers^{3,10} and swimmers¹¹ with complex body motions, and
43 robust jumpers^{6,12} have been developed based on this design strategy. However, in each case,
44 these robots are either tethered to or carry rigid systems for power and control, yielding hybrid
45 soft-rigid systems.^{4,8,10–12}

46 Creating a new class of fully soft, autonomous robots²⁵ is a grand challenge, as it requires
47 soft analogs of the control and power hardware currently used. Recently, monopropellant fuels
48 have been suggested as a promising fuel source for pneumatically actuated soft robots.^{4,14} Their
49 rapid decomposition into gas upon exposure to a catalyst offers a strategy for powering soft
50 robotic systems that obviates the need for batteries or external power sources. Here, we report a
51 method for creating a completely soft, pneumatic robot, the “octobot”, with eight arms that are
52 powered by monopropellant decomposition. To accomplish this, we use microfluidic logic¹³ as a
53 soft controller and multi-material, embedded 3D (EMB3D) printing method to fabricate

54 pneumatic networks within a molded elastomeric, robot body. Our hybrid assembly approach
55 allows one to seamlessly integrate soft lithography, molding, and 3D printing to rapidly and
56 programmably fabricate myriad materials and functional elements in form factors required for
57 autonomous, untethered operation of a soft robot.

58 To fabricate an octobot, we first micro-mold^{13,26} the soft controller that houses the
59 microfluidic logic necessary for controlling fuel decomposition (Figure 1a). The soft controller is
60 placed into a mold partially filled by hyperelastic layers needed for actuation (Figure 1b). Matrix
61 materials are then poured into the mold (Figure 1c), and the remaining soft robot features are
62 EMB3D printed into the molded matrix (Figure 1d, 1e, Supporting Video 1). After the matrix
63 materials are crosslinked, the aqueous fugitive inks “auto-evacuate” at elevated temperature as
64 water evaporates from the inks and diffuses through the matrix, leaving behind an open network
65 of channels that are interfaced with the soft controller (Figure 1f). Octobot fabrication is
66 completed upon removal of excess matrix material (Figure 1g). A more detailed description of
67 this multi-step assembly process is provided in Extended Data Figure 1.

68 By combining micro-molding with EMB3D printing, we rapidly patterned the required
69 mesofluidic networks by extruding sacrificial inks through fine nozzles that are embedded within
70 the uncured elastomer matrices. To self-heal crevices that form within the “body” matrix as the
71 nozzle is translated during the printing process, we created a new elastomeric material that
72 exhibits thixotropic behavior²⁷ (Extended Data Figure S2a). When completely restructured or at
73 rest, this matrix behaves like a Herschel-Bulkley fluid, i.e., it exhibits both shear-thinning
74 behavior (Extended Data Figure 2b), and a shear yield-stress (Extended Data Figure 2c). These
75 properties ensure that the extruded inks remain in place within the matrix.^{15,16} However, upon
76 yielding, the body matrix readily flows (Extended Data Figure 2c) into any crevices formed. The

77 body matrix restructures with time, ultimately recovering its original viscoelasticity (Extended
78 Data Figure 3), which ensures that EMB3D printing can be repeated later in the same matrix
79 region. We also created a “fuel reservoir” elastomeric matrix, into which fuel reservoir channels
80 are printed. Both the body and fuel reservoir matrices are crosslinked within the mold after
81 printing is completed.

82 To create the fuel reservoirs, catalytic reaction chambers, actuator networks, and vent
83 orifices, two hydrogel-based inks (fugitive and catalytic) are printed into the molded matrix
84 materials (Figure 2a). These printed features are interfaced with each other as well as the soft
85 controller through the use of “fugitive plugs” introduced at the controller’s inlets prior to filling
86 the mold with the matrix materials. The fugitive ink is composed of an aqueous, poly(ethylene
87 oxide)-*b*-poly(propylene oxide)-*b*-poly(ethylene oxide) triblock copolymer (Pluronic F127)
88 gel.^{15,28} The catalytic ink contains platinum particles (Supporting Video 2) suspended in a
89 mixture of Pluronic F127-diacrylate (F127-DA) and poly(ethylene glycol) diacrylate (PEG-DA)
90 that is photocrosslinked after printing. The rheological properties of both inks are specifically
91 tailored for EMB3D printing^{15,16} (Figure 2b, Extended Data Figure 4). The printed features
92 produced from both inks can be changed “on-the-fly” by varying the print speed (Figure 2c).
93 Typically, this fugitive ink must be removed or “evacuated” after printing to yield open
94 channels.^{15,28} However, we find that the fugitive ink composed of pure Pluronic F127 can be
95 auto-evacuated by heating the printed features within the crosslinked, silicone-based matrices at
96 90°C²⁹ (Figure 2d, Extended Data Figure 5). As water evaporation ensues, the triblock
97 copolymer species either form a thin coating at the matrix-open channel interface or they may
98 partially diffuse into the matrix. The fugitive plugs within the soft controller’s inlets also
99 undergo this auto-evacuation process, facilitating connectivity between the microfluidic logic

100 and all printed mesofluidic components (Extended Data Figure 6). By contrast, the catalytic ink
101 is crosslinked in place after printing yielding a Pt-laden plug within the matrix (Figure 2e).

102 To achieve the desired autonomous function, we incorporated a soft, microfluidic
103 controller within the octobot (Figure 3a). The control system is roughly divided into four
104 sections, upstream (liquid fuel storage), oscillator (liquid fuel regulation), reaction chamber
105 (decomposition into pressurized gas), and downstream (gas distribution for actuation and
106 venting). Upstream, 0.5 mL of fuel is infused via a syringe pump into each of two fuel reservoirs
107 printed into the hyperelastic matrix. Upstream check valves in the soft controller prevent fuel
108 from flowing back out the fuel inlets. The fuel reservoirs expand elastically to a pressure of
109 approximately 50 kPa, forcing fuel into the oscillator. The oscillator includes a system of pinch
110 and check valves based on prior designs,¹³ which convert pressurized fuel inflow into alternating
111 fuel outflow. With one channel temporarily occluded, fuel from the other channel flows from the
112 soft controller's outlets into the Pt laden reaction chambers, where it rapidly decomposes. The
113 resulting pressurized gas, prevented from returning to the soft controller via downstream check
114 valves, flows into one of the downstream mesofluidic networks comprised of four actuators and
115 one orifice. The supplied pressure deflects the actuators and exhausts to atmosphere through the
116 vent orifice. Thus, for robust actuation and timely venting, a balance must be reached between
117 supply gas flow, actuation pressure, and exhaust rate. These subcomponents operate based on the
118 interaction and timing of the local pressures, similar in concept to an electrical oscillator (Figure
119 3b). Upon successful venting, the fuel flow into one reaction chamber stops and flow to the other
120 begins, initiating a similar sequence in the other downstream catalytic chamber and actuator
121 network (Figure 3c).

122 To provide an on-board power source, we used a 50 wt% aqueous hydrogen peroxide as
123 the fuel due to its high energy density (1.44 kJ/g as compared to 0.1-0.2 kJ/g for batteries) as
124 well as its benign decomposition byproducts. As the fuel decomposes in the presence of the
125 platinum catalyst, the following reaction occurs $2\text{H}_2\text{O}_2 (\text{l}) \rightarrow 2\text{H}_2\text{O} (\text{l,g}) + \text{O}_2(\text{g})$, which results
126 in a volumetric expansion of approximately 240 times (at ambient pressure).¹⁴ At our operating
127 pressure of 50 kPa gauge, an expansion of 160 times is expected. Although higher fuel
128 concentrations would provide increased expansion and energy density, concentrations above 50
129 wt% drastically increase the decomposition temperature resulting in combustion of matrix
130 materials that surround the reaction chambers. Since this monopropellant liquid fuel can be
131 handled in small volumes and decomposes at the point of use, we can use microfluidic logic to
132 directly handle the fuel, eliminating the need for external valves¹⁰ to control gas at high pressure
133 and flow rate.

134 The geometry of the microfluidic soft controller is designed to operate at a fuel flow rate
135 of $\sim 40 \mu\text{L}/\text{min}$ thereby yielding pressurized gas at a rate of $\sim 6.4 \text{ mL}/\text{min}$.¹³ Under these
136 operating conditions, the theoretical runtime of 12.5 min could be achieved using a system with a
137 fuel capacity of 1 mL. The actuators, which consist of printed bladders in contact with a lower
138 modulus, hyperelastic elastomer layer (Figure 4a), are designed to inflate asymmetrically to
139 generate angular displacement. Their maximum working pressure and displacement are tuned
140 based on the thickness of the hyperelastic layer (Figure 4b, Extended Data Figure S7). If this
141 layer is too thin, it ruptures prematurely. However, the working pressure increases with
142 thickness. As a compromise, we selected a layer thickness of 1000 μm , as it affords consistent
143 performance at the lowest working pressure. In parallel with the actuators, we tailored the
144 diameter of the vent orifices by modulating print speed. Orifices roughly 75 μm in width allowed

145 proper actuator displacement with timely subsequent venting. The ability to rapidly pattern and
146 adjust the geometry of these features “on-the-fly” via EMB3D printing allowed us to iterate
147 through more than 30 designs and nearly 300 octobots to converge on an appropriate system-
148 level architecture.

149 Through this iterative process, we created octobots with embedded components that work
150 together in concert to alternate between the red and blue actuation states shown in Figure 4c. The
151 resulting octobots operated autonomously (Figure 4d, Supplemental Video 3), cycling between
152 actuation states for four to eight minutes. While this is less than the predicted theoretical runtime,
153 the soft controller alternates actuation states as expected. We believe that downstream
154 impedances arising from decomposition-actuation-venting cycles as well as the decreasing flow
155 rate of fuel into the soft controller with time are responsible for the departure from theoretical
156 performance.¹³ Further advances in microfluidic logic design for soft robotic control will lead to
157 longer periods of actuation cycles, account for dynamic fuel input, and also facilitate more
158 sophisticated gait cycles that will enable true locomotion.

159 In summary, we have demonstrated the first untethered operation of a robot composed
160 solely of soft materials. The coupling of monopropellant fuels and microfluidic logic allowed us
161 to power, control, and realize autonomous operation of these pneumatically actuated systems.
162 Through our hybrid assembly approach, we constructed both the robot body and embedded the
163 necessary components for fuel storage, catalytic decomposition, and actuation to enable system-
164 level function in a rapid manner. The octobot is a minimal system designed to demonstrate our
165 integrated design and fabrication strategy, which may serve as a foundation for a new generation
166 of completely soft, autonomous robots.

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168 Methods**169 Soft Controller Fabrication**

170 Soft controllers are fabricated from Sylgard 184 PDMS (Dow Corning Corp. Auburn, MI, USA)
171 using soft lithography molding and bonding techniques. First, a mold was fabricated on a silicon
172 wafer using SU-8 negative photoresist (Microchem, Corp. Westborough, MA, USA). SU-8 3050
173 photoresist was used to achieve 100 nm film thickness. Baking, exposing, and developing steps
174 were performed in accordance with product specifications in the product datasheet. The
175 completed wafer is placed in a petri dish to form a completed mold assembly.

176 Soft controllers consist of an upper mold, a lower mold, and an intermediate thin film.
177 The upper and lower molds are made on one wafer to ease fabrication. PDMS is poured into the
178 mold assembly to a height of one millimeter. Separately, PDMS is spun coat onto a wafer at
179 1500 rpm for 60 seconds for a film thickness of 35 nm. After curing at 90° C for 20 minutes,
180 PDMS forms are removed from the molds, and holes are punched at all inlets and outlets. The
181 upper layer is bonded to the wafer-adhered thin film after exposing to oxygen plasma at 35 Watts
182 for 20 seconds in a Deiner Pico plasma system (Deiner Electronic GmbH). Holes are punched in
183 the thin film, masks are placed as described by Mosadegh, et al.,²⁶ and the lower layer is bonded
184 to the thin film using the plasma recipe above.

185 Ink and Matrix Materials

186 Two inks, a “fugitive ink” and “catalytic ink,” are formulated for EMB3D printing. The fugitive
187 ink is prepared by adding 27 wt% gel of Pluronic F127 to ice-cold, deionized, ultra-filtrated
188 (DIUF) water, followed by mixing in a planetary mixer for 5 min at 2000 rpm, and storing at
189 4°C. The fugitive ink is not used until the Pluronic F127 completely dissolves in solution. The
190 ink is prepared for printing by loading the solution at 4°C in a 3 cc syringe barrel (EFD Nordson,

191 East Providence, RI, USA) and centrifuged at 3000 rpm for 5 min to degas. For EMB3D
192 printing, the fugitive ink's barrel is fitted with a stainless steel nozzle (0.15 mm inner diameter,
193 EFD Nordson).

194 The catalytic ink is prepared by first synthesizing and then dissolving a diacrylated
195 Pluronic (F127-DA) at 30wt% concentration with a solution of Irgacure 2959 (at 0.5wt%, BASF)
196 in DIUF water at 4°C. The F127-DA is synthesized under an inert nitrogen atmosphere by first
197 adding 400 mL of dry toluene (Sigma, St. Louis, MO, USA) to a three-neck flask fixed to a
198 condenser with circulating cold water and magnetically stirred at 300 rpm. 70 g of Pluronic F127
199 (Sigma) is then dissolved in the toluene after heating the solvent to 60°C. After the solution is
200 allowed to cool to room temperature, triethylamine (5.6 g, Sigma) is added to the solution,
201 followed by the drop-wise addition of acryloyl chloride (5 g, Sigma) with continued stirring,
202 both at a molar ratio of 10:1 with the Pluronic F127. The reaction mixture is stirred overnight
203 and maintained in the inert atmosphere. The diacrylated Pluronic F127 (F127-DA) product is
204 then filtered from the yellow triethylammonium hydrochloride byproduct and precipitated from
205 the filtered solution with hexane (Sigma) at a 1:1 volume ratio. The F127-DA is obtained
206 through a second filtration step and allowed to dry in a chemical hood for at least 24 h. This
207 protocol is adapted from Wu, et al.¹⁵ For each gram of this base F127-DA mixture, 100 mg of
208 PEG-DA is added, and this solution is mixed in a planetary mixer for 1 min at 2000 rpm and
209 degassed for 3 min at 2200 rpm. This mixture is then stored in the dark at 4°C. Finally, 5 w/w%
210 Pt black (Sigma) is added to this base solution at 4°C and mixed in a planetary mixer for 5 min at
211 2000 rpm. The Pt-filled F127-DA physically gels during mixing, facilitating loading into a UV-
212 blocking 3cc syringe barrel (EFD Nordson) for printing. Note, this catalytic ink is freshly
213 prepared for each print session, as the Pt black slowly crosslinked the acrylate moieties present in

214 the ink. After EMB3D printing, the catalytic ink is crosslinked for 15 min at 18 mW/cm^2 under a
215 UV source (Omniculture EXFO). For EMB3D printing, the syringe barrel housing this ink is fitted
216 with a stainless steel nozzle (0.33 mm inner diameter, EFD Nordson).

217 Two matrix materials are developed for fabricating fully soft robots. The first matrix,
218 referred to as the “body matrix,” is prepared by blending two silicone-based materials: Sylgard
219 184 and SE 1700 (Dow Corning). Sylgard 184 PDMS is used to dilute SE 1700 to achieve the
220 desired rheological response for embedded 3D printing. After exploring several blends, we find
221 that the optimal body matrix is composed of a 1:1 mass ratio of SE 1700 (4:1 ratio of base to
222 hardener) and Sylgard 184 (10:1 ratio of base to hardener). This matrix is prepared by mixing the
223 blend in a planetary mixer at 2000 rpm for 3 min with degassing at 2200 rpm for 2 min. The
224 second matrix, referred to as the “fuel reservoir matrix,” is prepared by mixing Part A Ecoflex
225 00-30 to Part B Ecoflex 00-30 (with 1.2 w/w% Slo-Jo Platinum Silicone Cure Retarder and 1.2
226 w/w% Thivex, Smooth-On Inc., Macungie, PA, USA) in a 1:1 ratio. The matrix is prepared in a
227 planetary mixer at 2000 rpm for 1.5 min with degassing at 2200 rpm for 1 min.

228 Lastly, the “fugitive plug” material used to prevent ingress of the body matrix material
229 into the soft controller is prepared prior to printing by first synthesizing and then mixing a
230 diacrylated Pluronic material (F127-DA) (at 30 wt% in a 0.5 wt% solution of Irgacure 2959 in
231 DI water) with F127 (at 30 wt% in DI water) at a mass ratio of 1:4. The fugitive plug is stored in
232 the dark at 4°C in a syringe. When used, the fugitive plug material is allowed to physically gel
233 before it is crosslinked for 3 min at 6 mW/cm^2 under a UV source.

234 **Rheological Characterization**

235 All rheological measurements are carried out using a controlled-stress rheometer (DHR-3, TA
236 Instruments, New Castle, DE, USA) equipped with a 40 mm diameter, 2° cone and plate

237 geometry. In all experiments, the fugitive and catalytic inks are equilibrated at room temperature
238 for 1 min before testing; the fuel reservoir and body matrix materials are equilibrated for 20 min
239 and 10 min, respectively, to simulate the times at which octobot printing began with each
240 material. Shear storage moduli are measured as a function of shear stress at a frequency of 1Hz.

241 The body matrix materials are characterized by both sweep and flow tests to determine
242 their rheological response (Extended Data Figure 2). In addition, three-phase modulus recovery
243 tests are carried out to quantify the recovery time of the body matrix stiffness after applying a
244 shear stress that exceeds the equilibrium yield stress, $\tau_{y,0}$ (Extended Data Figure 3). In the first set
245 of experiments, flow sweeps from low (10^{-2} s^{-1}) to high (10^2 s^{-1}) shear rates are carried out and
246 immediately followed by ramp sweeps from high to low shear rates. In the latter set, of
247 experiments, shear storage (G') and loss (G'') moduli are measured during three phases of applied
248 shear stresses (at 1 Hz frequency): 1 Pa for 3 min; 100 Pa for either 1, 10, or 100s; and 1 Pa for
249 30 min. We defined their thixotropic recovery time, as the instant $G' = G''$, or when $\tan(\delta) = G''/$
250 $G' = 1$.

251 **Actuator Characterization.** Actuators are printed into special actuator characterization molds
252 by EMB3D printing and then auto-evacuate. To prepare them for characterization, they are first
253 released from mold assembly and then a 1 mm hole is created with a biopsy punch (Miltex Inc.
254 York, PA, USA), which serves as the air inlet. Finally, the actuator is pressurized slightly to
255 insure inflation. Each actuator design is tested for blocked force (i.e., the actuator is constrained
256 from deflection and resultant force is measured) and free displacement (i.e., the actuator is
257 allowed to deflect unconstrained and the total displacement is measured). For blocked force
258 characterization, individual actuators are mounted on a fixed platform beside an Instron model
259 5544 materials testing frame (Illinois Tool Works Inc., Norwood, MA, USA). The actuator is

260 lowered until just above the force sensor portion of the testing frame, and the actuator is plumbed
261 with regulated compressed air. Actuators typically behave differently upon initial few actuations
262 versus subsequent actuations due to the Mullins effect.³⁰ Each actuator therefore receives five
263 “break in” cycles prior to data acquisition.

264 For each actuator, break in testing consists of five cycles, in which actuator air pressure is
265 slowly (~30 sec) ramped up to the pressure set point, then slowly (~30 sec) ramped down to
266 ambient. Pressure set point for the first cycle is P_0 , and set point for all following cycles is P_I
267 (Table S1). Data acquisition consists of five additional cycles for each actuator, in which air
268 pressure is cycled as above to pressure set point P_I . Air pressure and actuator force data is
269 recorded on the Instron testing frame data acquisition system at 100 ms intervals.

270 For free displacement characterization, actuators are plumbed with regulated compressed
271 air, and mounted vertically between a matte black background and a Sony NEX3 digital camera
272 for video data acquisition. Actuators are pressurized with five break-in cycles as described
273 above, followed by five data acquisition cycles. As above, the first break in cycle is to P_0 , and all
274 subsequent break in and data acquisition cycles are to P_I . Video data is analyzed using ImageJ
275 image analysis platform (NIH.gov) to obtain bend angle versus pressure for each actuator.

276 **Mold Fabrication**

277 Octobot molds are fabricated inside a CNC machined Delrin® mold equipped with two
278 locating pins to mount the soft controller. Their desired shape is modeled in Solidworks. A
279 negative mold is modeled, and output in Parasolid format for file transfer. MasterCAM is used to
280 develop all machining tool paths and to export final G-code for final fabrication. Blanks of 105
281 mm length were cut from black acetal (Delrin®), stock size 1x3 inch (McMaster Carr, Santa Fe
282 Springs, CA, USA). Acetal is used due to its dimensional stability, and thick stock is chosen to

283 prevent warping during machining and repeated octobot curing cycles. Octobot molds are
284 produced by CNC milling on a HAAS OM-2A vertical machining center (HAAS Automation
285 Inc, Oxnard, CA, USA). 1 mm dowel pins are pressed into drilled holes for controller mounting.

286 **Soft Robot Assembly**

287 A custom-designed, multi-material 3D printer (ABL 10000, Aerotech Inc., Pittsburg, PA,
288 USA) with four independently z-axis addressable ink reservoirs is used to pattern fugitive and
289 catalytic inks within the octobot matrices.²⁸ All G-Code for printing is generated from Python-
290 based software (MeCode, developed by J. Minardi). Prior to EMB3D printing, Ecoflex 30
291 (Smooth-On, Inc.) is first prepared with 1 wt% Slo-Jo and 0.25 wt% Thivex (both with respect to
292 Part A) by mixing in a Thinky planetary mixer for 1.5 min with a 1 min degas cycle. This
293 uncured Ecoflex 30 is cast into the actuator layers of the octobot mold and degassed in a vacuum
294 chamber for 3 min. A glass slide is used to remove excess material and create smooth surfaces
295 that will ultimately become the extensible layers of the actuators. The molds are then placed in a
296 90°C oven for 30 min to cure the Ecoflex, removed, and trimmed of excess material as
297 necessary.

298 A soft controller is then loaded onto the press-fit pins placed in the printing mold with the
299 Kapton tape still adhered. Registration coordinates and print heights are then taken from the
300 cured Ecoflex layers in the actuators and in all inlets of the soft controller; these are essential for
301 EMB3D printing and provided to the custom print software. The fuel reservoir and body matrix
302 materials are prepared as described previously. While the body matrix material is mixing, the
303 fuel reservoir matrix is deposited in the fuel reservoir region of the printing mold and degassed
304 for three minutes. Excess bubbles in the uncured fuel reservoir matrix are removed with a
305 pipettor. Non-gelled, chilled fugitive plug is then filled throughout the soft controller via

306 injection through the inlets. While the fugitive plug is still in the liquid state, it is briefly
307 degassed in a vacuum chamber. The fugitive plug material is then allowed to physically
308 crosslink, excess gel is scraped from the top of the tape, the tape is removed, and the fugitive
309 plug is photocrosslinked with a UV source at 6 mW/cm^2 for 3 min. After the gels are
310 crosslinked, the body matrix is cast within the mold, covering the fuel reservoir matrix and the
311 fugitive plug-filled soft controller and degassed for 1-3 min. Again, excess bubbles are removed
312 with a pipettor, excess material is scraped off and away from the mold with a glass slide, and
313 EMB3D printing of the fugitive and catalytic inks begins. After printing, the entire mold is cured
314 at 18 mW/cm^2 for 15 min to crosslink the catalytic ink. The mold is then transferred to a 90°C
315 oven, where the matrix materials crosslink. The octobot is removed from the mold and kept at
316 90°C for 4 days to facilitate auto-evacuation of the inks.

317 After auto-evacuation, the octobot is release cut from the surrounding matrix material
318 using a CO_2 laser (Universal Laser Systems, Scottsdale, AZ, USA) and cleaned with isopropyl
319 alcohol and water. Sylgard 184 PDMS (Dow Corning Corp. Auburn, MI, USA) is poured into
320 the octobot's open cavity above the soft controller to a height of 1.5 mm and cured at 90°C for
321 20 min. A 1 mm biopsy punch (Militex Inc, York, PA, USA) is used to punch holes through the
322 newly poured PDMS layer and into the fuel inlets. Dyed water is injected into these holes to
323 inflate the fuel tanks, flow through the system, and insure proper bot function. Holes are punched
324 in the downstream orifice features to allow the water to vent from the system.

325 The octobot is loaded into an acrylic tank outfitted with a backlight to highlight colored
326 fuel as it flows through the system. Aqueous hydrogen peroxide (90 wt%, HTP grade,
327 Peroxychem, Philadelphia, PA, USA) is diluted to 50 wt% and samples dyed red and blue are
328 filled into two syringes prepared with this liquid fuel mixture. The syringes are loaded onto a

329 syringe pump, and connected to the octobot via 1mm diameter silicone rubber tubing. Water is
330 flowed into the acrylic tank to wash away dye in the octobot exhaust stream and drained into a
331 nearby sink. The syringe pump flows fuel at a rate of 3 mL/min (each syringe) into the octobot
332 for 10 sec. The silicone rubber tubing is removed with tweezers from the octobot, which is
333 allowed to operate untethered. The octobot alternates actuation until fuel pressure is insufficient
334 to switch the oscillator and alternating actuation ceases.

335 **Imaging and Videography**

336 Photographs and supporting videos are acquired with a digital SLR camera (Canon EOS
337 5D Mark II, Canon USA Inc) and a 4K video (Blackmagic Production 4K, Blackmagic Design,
338 Melbourne, Australia). Photos are cropped using Inkscape vector graphics editor
339 (www.inkscape.org), and video sequences are clipped from raw footage and exported using
340 iMovie (Apple Corp, Cupertino, CA, USA). All print parameter measurements and images of
341 EMB3D printed features in octobots are taken with a digital zoom microscope (VHX-2000,
342 Keyence, Japan). Their mean values and standard deviations are determined from three samples
343 printed at each print speed of interest.

344

345 **References**

- 346 1. Rus, D. & Tolley, M. T. Design, fabrication and control of soft robots. *Nature* **521**, 467–
347 475 (2015).
- 348 2. Wang, L. & Iida, F. Deformation in Soft-Matter Robotics: A Categorization and
349 Quantitative Characterization. *IEEE Robot. Autom. Mag.* **22**, 125–139 (2015).
- 350 3. Shepherd, R. F. *et al.* Multigait soft robot. *Proceedings of the National Academy of*
351 *Sciences* **108**, 20400–20403 (2011).

- 352 4. Onal, C. D., Chen, X., Whitesides, G. M. & Rus, D. Soft mobile robots with on-board
353 chemical pressure generation. in *International Symposium on Robotics Research (ISRR)*
354 1–16 (2011).
- 355 5. Lin, H.-T., Leisk, G. G. & Trimmer, B. GoQBot: a caterpillar-inspired soft-bodied rolling
356 robot. *Bioinspir. Biomim.* **6**, 026007 (2011).
- 357 6. Shepherd, R. F. *et al.* Using explosions to power a soft robot. *Angew. Chemie - Int. Ed.* **52**,
358 2892–2896 (2013).
- 359 7. Martinez, R. V. *et al.* Robotic tentacles with three-dimensional mobility based on flexible
360 elastomers. *Adv. Mater.* **25**, 205–212 (2013).
- 361 8. Stokes, A. A., Shepherd, R. F., Morin, S. A., Ilievski, F. & Whitesides, G. M. A Hybrid
362 Combining Hard and Soft Robots. *Soft Robot.* **1**, 70–74 (2014).
- 363 9. Mosadegh, B. *et al.* Pneumatic Networks for Soft Robotics that Actuate Rapidly. *Adv.*
364 *Funct. Mater.* **24**, 2163–2170 (2014).
- 365 10. Tolley, M. T. *et al.* A Resilient, Untethered Soft Robot. *Soft Robot.* **1**, 213–223 (2014).
- 366 11. Marchese, A. D., Onal, C. D. & Rus, D. Autonomous Soft Robotic Fish Capable of
367 Escape Maneuvers Using Fluidic Elastomer Actuators. *Soft Robot.* **1**, 75–87 (2014).
- 368 12. Bartlett, N. W. *et al.* A 3D-printed, functionally graded soft robot powered by combustion.
369 *Science (80-.).* **349**, 161–165 (2015).
- 370 13. Mosadegh, B. *et al.* Integrated elastomeric components for autonomous regulation of
371 sequential and oscillatory flow switching in microfluidic devices. *Nature Physics* **6**, 433–
372 437 (2010).
- 373 14. Wehner, M. *et al.* Pneumatic Energy Sources for Autonomous and Wearable Soft
374 Robotics. *Soft Robot.* **2**, 141031124812001 (2014).

- 375 15. Wu, W., Deconinck, A. & Lewis, J. A. Omnidirectional printing of 3D microvascular
376 networks. *Adv. Mater.* **23**, H178–83 (2011).
- 377 16. Muth, J. T. *et al.* Embedded 3D printing of strain sensors within highly stretchable
378 elastomers. *Adv. Mater.* **26**, 6307–6312 (2014).
- 379 17. Palleau, E., Morales, D., Dickey, M. D. & Velev, O. D. Reversible patterning and
380 actuation of hydrogels by electrically assisted ionoprinting. *Nat. Commun.* **4**, 2257 (2013).
- 381 18. Ionov, L. Biomimetic hydrogel-based actuating systems. *Adv. Funct. Mater.* **23**, 4555–
382 4570 (2013).
- 383 19. Anderson, I. A., Gisby, T. A., McKay, T. G., O’Brien, B. M. & Calius, E. P. Multi-
384 functional dielectric elastomer artificial muscles for soft and smart machines. *J. Appl.*
385 *Phys.* **112**, 0–20 (2012).
- 386 20. Brown, E. *et al.* Universal robotic gripper based on the jamming of granular material.
387 *Proc. Natl. Acad. Sci.* **107**, 18809–18814 (2010).
- 388 21. Ilievski, F., Mazzeo, A. D., Shepherd, R. F., Chen, X. & Whitesides, G. M. Soft robotics
389 for chemists. *Angew. Chemie - Int. Ed.* **50**, 1890–1895 (2011).
- 390 22. Shepherd, R. F., Stokes, A. A., Nunes, R. M. D. & Whitesides, G. M. Soft machines that
391 are resistant to puncture and that self seal. *Adv. Mater.* **25**, 6709–6713 (2013).
- 392 23. Tolley, M. T. *et al.* A Resilient, Untethered Soft Robot. **1**, 213–223 (2014).
- 393 24. Mosadegh, B. *et al.* Pneumatic Networks for Soft Robotics that Actuate Rapidly. *Adv.*
394 *Funct. Mater.* **24**, 2163–2170 (2014).
- 395 25. Whitesides, G. M. What comes next? *Lab Chip* **11**, 191–193 (2011).
- 396 26. Mosadegh, B., Tavana, H., Lesher-Perez, S. C. & Takayama, S. High-density fabrication
397 of normally closed microfluidic valves by patterned deactivation of oxidized

- 398 polydimethylsiloxane. *Lab Chip* **11**, 738–742 (2011).
- 399 27. Barnes, H. A. Thixotropy - A review. *J. Nonnewton. Fluid Mech.* **70**, 1–33 (1997).
- 400 28. Kolesky, D. B. *et al.* 3D bioprinting of vascularized, heterogeneous cell-laden tissue
401 constructs. *Adv. Mater.* **26**, 3124–3130 (2014).
- 402 29. Randall, G. C. & Doyle, P. S. Permeation-driven flow in poly(dimethylsiloxane)
403 microfluidic devices. *Proc. Natl. Acad. Sci. U. S. A.* **102**, 10813–10818 (2005).
- 404 30. Mullins, L. Softening of Rubber by Deformation. *Rubber Chemistry and Technology* **42**,
405 339–362 (1969).

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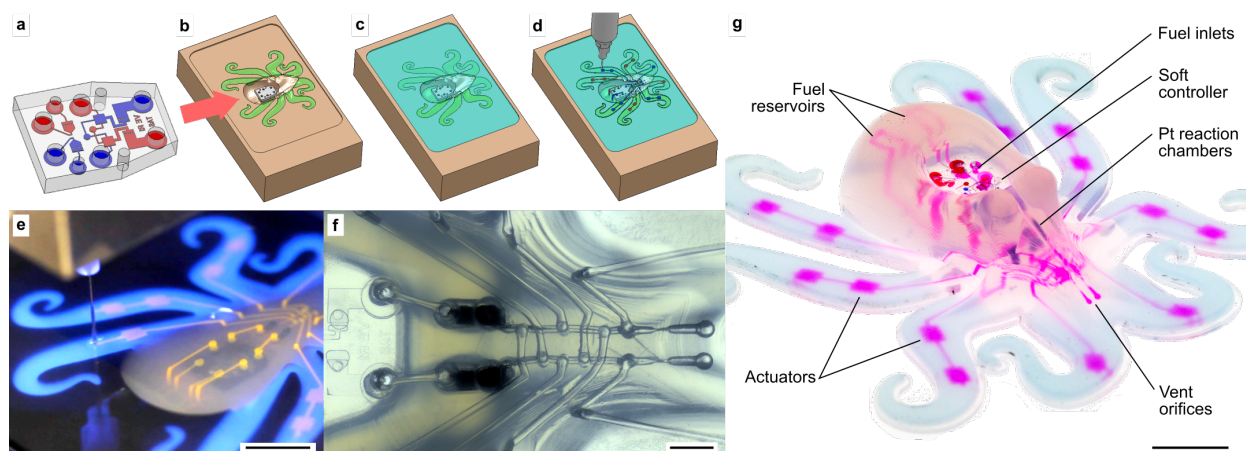
417 **Author Contributions**

418 M.W., R.L.T., J.A.L., and R.J.W. conceived the experimental work; M.W. and R.L.T. led the
419 experiments with assistance from D.J.F. and B.M; M.W., R.L.T., J.A.L., and R.J.W. contributed
420 to data analysis and interpretation and wrote the paper. All authors provided feedback.

421 **Author Information**

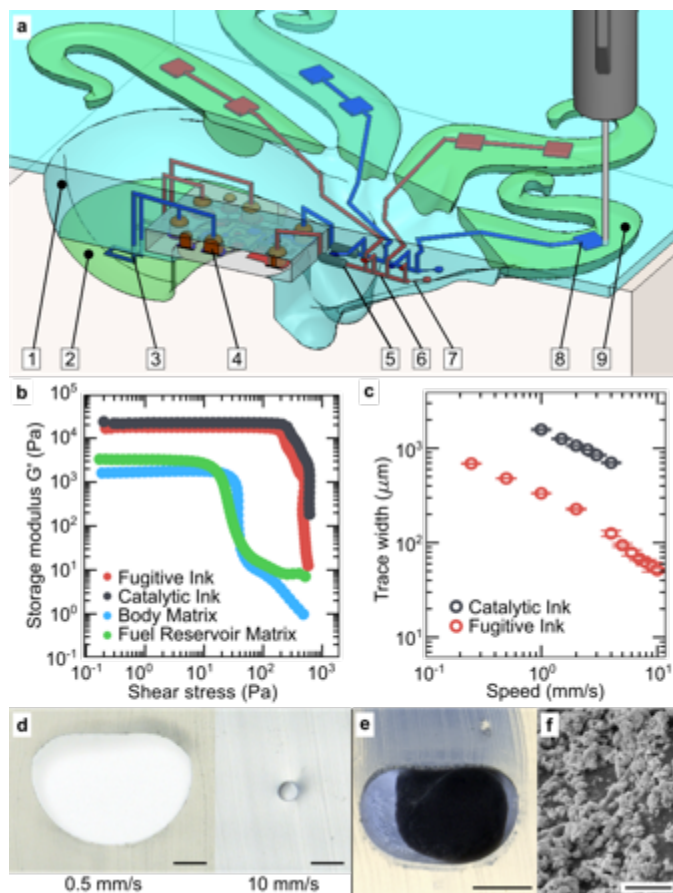
422 Reprints and permissions information is available at www.nature.com/reprints. The authors
423 declare no competing financial interests. Correspondence and requests for materials should be
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426 **Figures and Figure Legends**

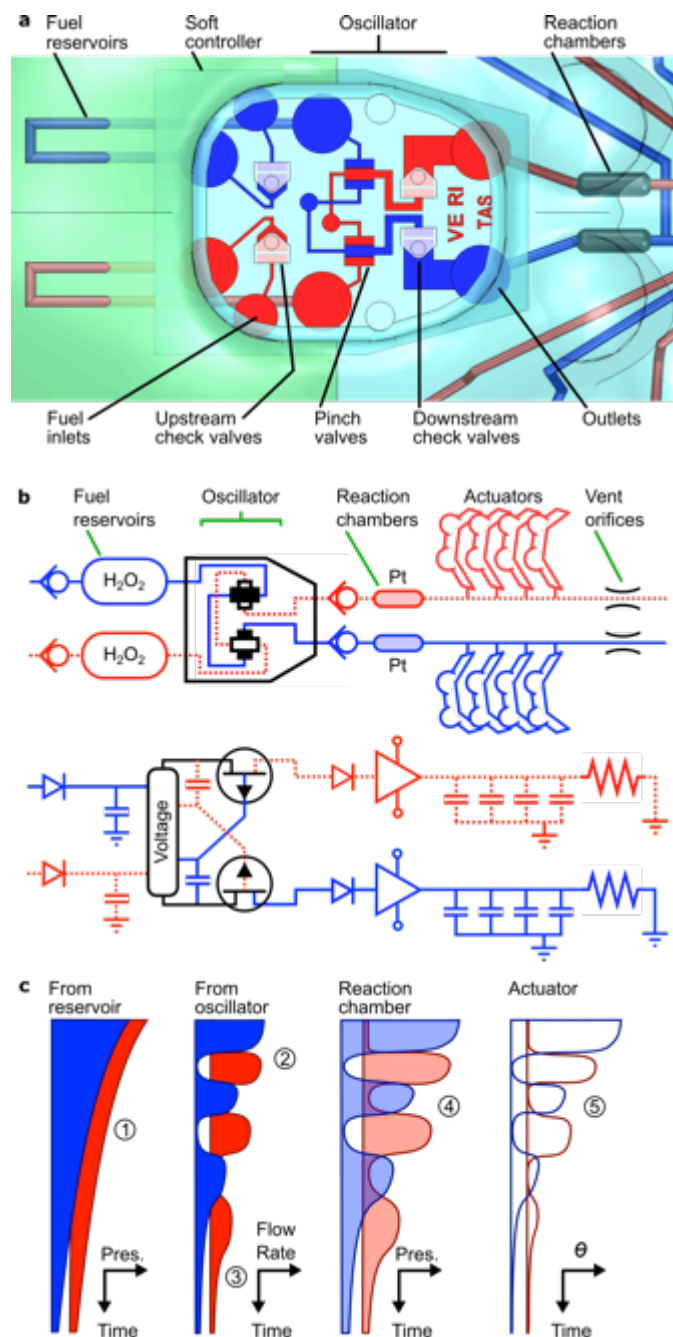
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 428 **Figure 1. Fully soft, autonomous robot assembly.** **a**, Pre-fabricate a microfluidic soft
 429 soft controller, and **b**, load this controller into a mold. **c**, Pour matrix materials into the mold and **d,e**,
 430 EMB3D print fugitive and catalytic inks (scale bar in **e** represents 10 mm). **f**, After matrix
 431 curing, the printed fugitive ink “auto-evacuates” yielding open channels (scale bar represents 2
 432 mm). **g**, After curing, the octobot is removed from the mold and inverted to reveal an
 433 autonomous, fully soft robot, controlled via the embedded microfluidic soft controller and
 434 powered by monopropellant decomposition (scale bar represents 10 mm). Fluorescent dyes have
 435 been added in **e** and **g** to assist in visualization of internal features.

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441 **Figure 2. Multimaterial, EMB3D printing.** **a**, The octobot features include (1) the body matrix,
442 (2) fuel reservoir matrix, (3) printed fuel reservoir traces, (4) fugitive plugs in soft controller, (5)
443 printed Pt reaction chamber, (6) printed pneumatic network, (7) printed vent orifices, (8) printed
444 actuators, and (9) molded hyperelastic actuator matrix. All printed features are composed of the
445 fugitive ink except (5), which is patterned using the catalytic ink. **b**, The storage modulus, G' , of
446 the fugitive ink, catalytic ink, body matrix, and fuel reservoir matrix is provided as a function of
447 shear stress. The plateau storage moduli of the inks are an order of magnitude higher than those
448 of the matrix materials. **c**, Trace widths of the fugitive and catalytic inks printed at 65 psi (450
449 kPa) and 50 psi (345 kPa), respectively, decrease with print speed (error bars indicate standard
450 deviation). **d**, Optical images of channel cross-sections printed at speeds of 0.5 and 10 mm/s,
451 which demonstrate that trace dimensions can be changed “on-the-fly” (scale bar is 100 μm).
452 Reaction chambers printed with the catalytic inks containing a Pt-laden plug, as shown in **e**, a
453 cross-section and **f**, scanning electron micrograph. (Scale bars in **e** and **f** represent 500 μm and
454 25 μm , respectively.)

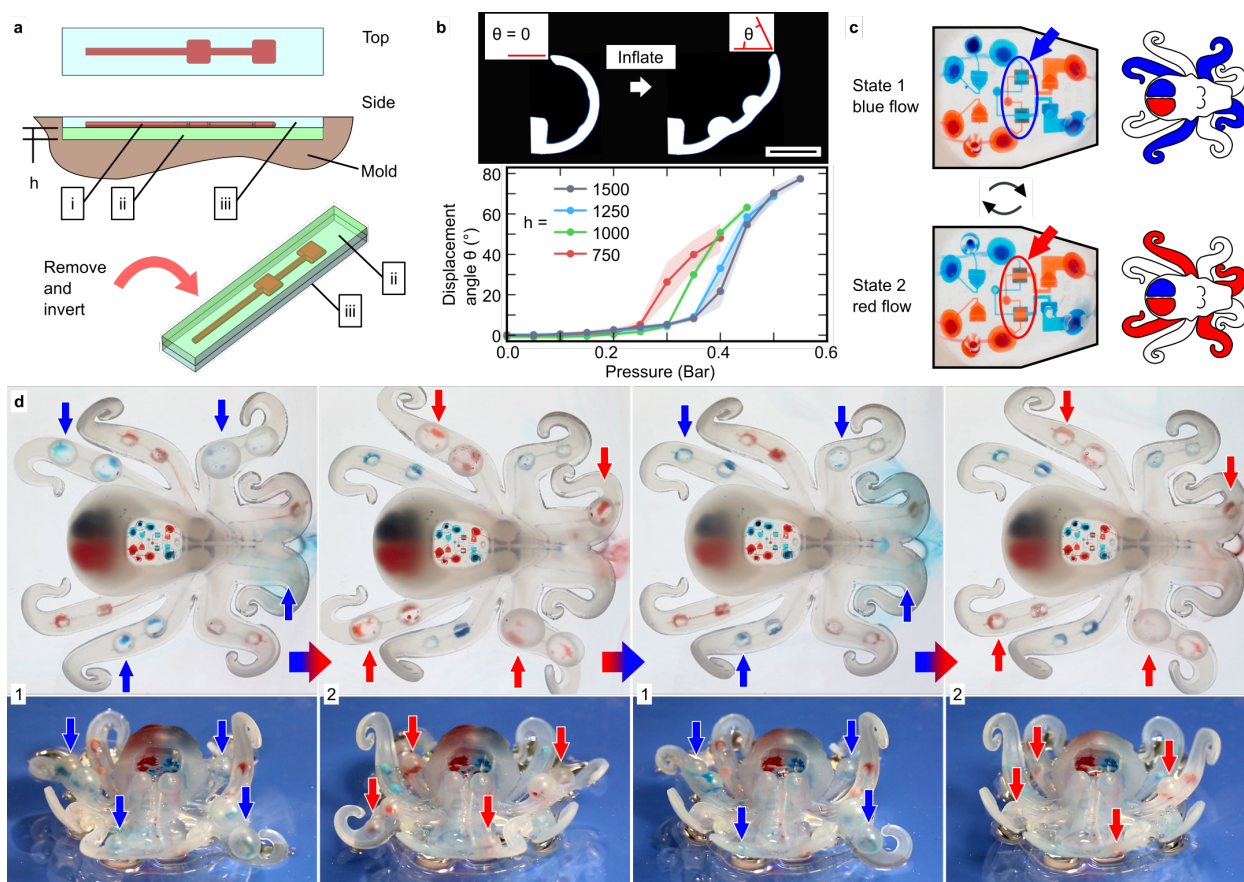


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Figure 3. Octobot control logic. Discrete sides are shown in red and blue for clarity. **a**, A system of check valves and switch valves within the soft controller regulates fluid flow into and through the system. **b**, A schematic and qualitative electrical analogy of the octobot system are provided, where check valves, fuel tanks, oscillator, reaction chambers, actuators, and vent orifices are akin to diodes, supply capacitors, electrical oscillator, amplifiers, capacitors, and pull down resistors, respectively. **c**, Conceptual curves show key variables as a function of time. (1) Nominal pressure drives fuel through system at a decreasing rate. (2) Pinch valves in the oscillator convert upstream flow into alternating flow between red and blue channels. Flow rate and switching frequency are functions of upstream pressure and downstream impedance. (3) When upstream pressure is too low, oscillation is not possible, so both sides flow at reduced rate.

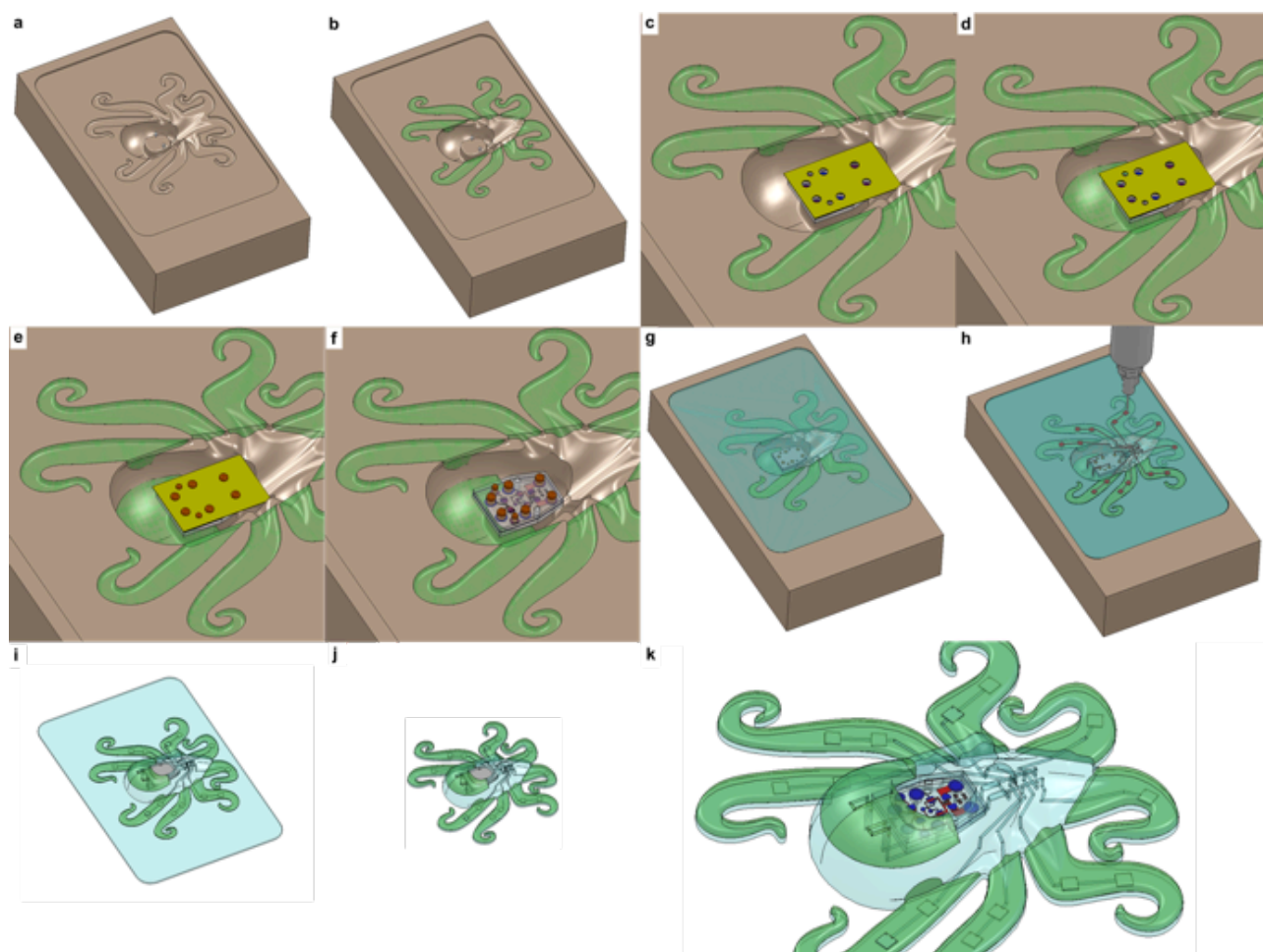
467 (4) Catalyst decomposes fuel, yielding pressurized gas, which flows downstream to the actuators
468 and the vent orifices concurrently. (5) Actuators deform based on pressure. Vents must be
469 sufficiently small to allow full actuation, yet sufficiently large to allow timely venting.

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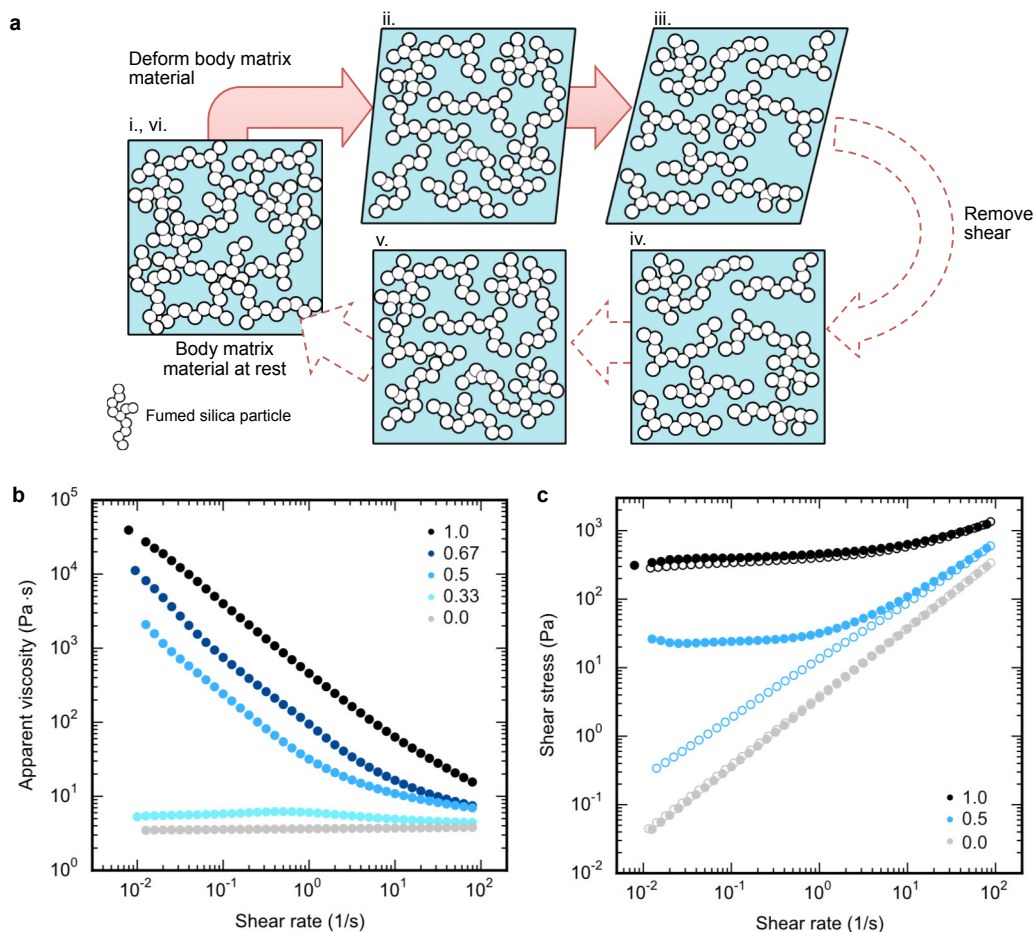
471
 472 **Figure 4. Octobot actuation.** **a**, Actuator design in which traces (i) are printed in contact with
 473 the hyperelastic layer (ii) inside of the body matrix material and (iii) differences in modulus
 474 result in bending upon inflation. The thickness, h , of the hyperelastic layer is modified to change
 475 actuator characteristics. In this example, the body matrix material (iii) possesses a height of 800
 476 μm . **b**, (Top) The actuator tip angle θ changes upon inflation (scale bar represents 10 mm).
 477 (Bottom) Displacement angle θ as a function of inflation pressure is provided for actuators with
 478 varying hyperelastic layer height, h , (in microns). Error bars indicate 95% confidence interval. **c**,
 479 The soft controller's oscillator causes an octobot to alternate between blue and red actuation
 480 states. The monopropellant fuel is dyed to show states. **d**, Stills from top-down (top) and face-on
 481 (bottom) operation videos show an octobot autonomously alternating between blue and red
 482 actuation states.

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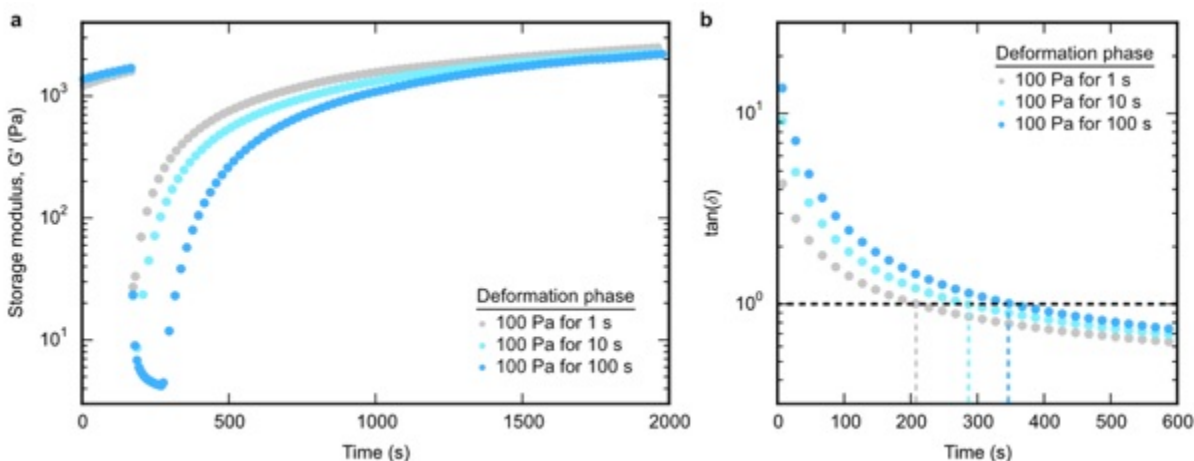
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485 **Extended Data Figure 1. Workflow for EMB3D printing an octobot.** **a**, An EMB3D printing
 486 mold is machined from Delrin®. **b**, The hyperelastic layers needed for actuation are cast and
 487 crosslinked in the mold's actuator regions. **c**, A soft controller protected with a polyimide tape
 488 mask is loaded onto the EMB3D printing mold's pins. **d**, The fuel reservoir matrix material is
 489 carefully loaded into the fuel reservoir area of the mold and degassed under vacuum. **e**, Liquified
 490 fugitive plug material is manually loaded into the soft controller via the inlets and briefly
 491 degassed. **f**, The protective tape is removed after the fugitive plug material physically gels, and
 492 the fugitive plug is photocrosslinked. **g**, The body matrix material is cast into the mold and
 493 degassed. **h**, Any excess body matrix material is removed with a squeegee step, EMB3D printing
 494 begins, and the entire mold and EMB3D printed materials are placed in a 90°C oven to crosslink.
 495 **i**, After two hours, the crosslinked octobot is removed from its mold and kept at 90°C for a total
 496 of four days to ensure complete auto-evacuation of the aqueous fugitive inks. **j**, Before operation,
 497 excess body matrix material is removed via laser cutting. **k**, The final octobot, shown here in a
 498 close-up view, is prepared for operation.

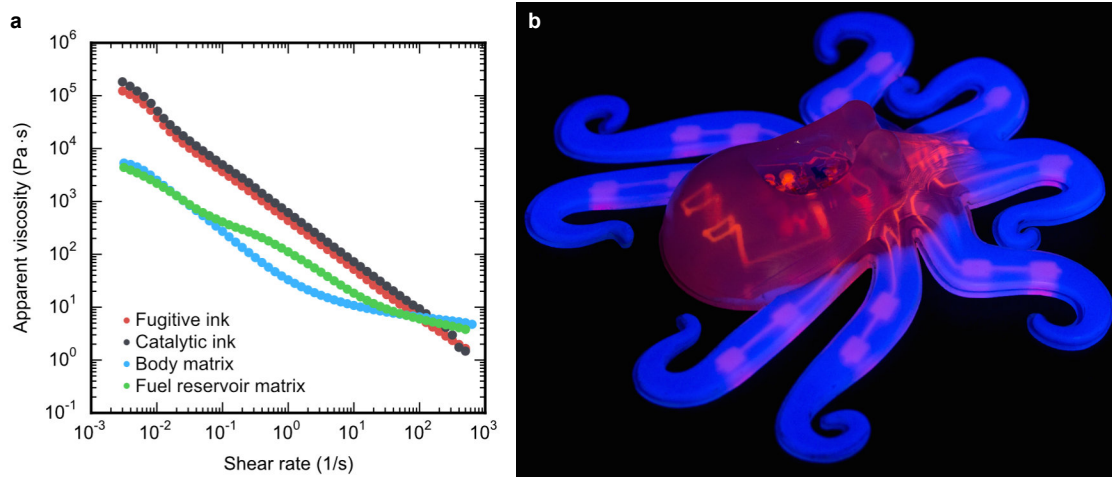


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 500 **Extended Data Figure 2. Rheological properties of the body matrix.** **a**, Schematic illustration
 501 of the body matrix behavior during the EMB3D printing process. (i) When the body matrix is at
 502 rest, the fumed silica fillers within the silicone material form a percolated network, which give
 503 rise to its shear yield stress, $\tau_{y,0}$. (ii) As nozzle travels through the matrix during printing, the
 504 matrix is yielded, and the percolated filler network is disrupted, which decreases the yield stress.
 505 (iii) Sufficient deformation can completely disrupt the fumed silica microstructure and
 506 completely eliminate the matrix material's yield stress ($\tau_{y,t} \rightarrow 0$ Pa). (iv) The fumed silica
 507 network does not immediately recover when it returns to a quiescent state. (v) Over time, the
 508 network slowly restructures to (vi) its equilibrium microstructure. **b,c**, Log-log plots of apparent
 509 viscosity (**b**) and corresponding shear stress versus shear rate (**c**) are shown for various PDMS
 510 matrix formulations investigated, which are prepared by blending Sylgard 184 (10:1 ratio of base
 511 to hardener) and SE 1700 (4:1 ratio of base to hardener) at various mass fractions. The
 512 formulations are listed by the weight ratio of SE 1700 used (0.0, 0.33, 0.5, 0.67, 1.0). **c**, Closed
 513 and open circles in the plot represent measurements taken during the flow sweep and ramp steps
 514 of the thixotropic loop studies, respectively. The final body matrix, formulated from the 50 wt%
 515 SE 1700 blend, shows clear thixotropic behavior and a significant decrease in yield stress upon
 516 yielding. Blends with higher concentrations of filler particles show diminished thixotropic
 517 behavior, and the yield stress is not eliminated during nozzle translation. Consequently, crevices
 518 or air pockets form during printing with matrix materials possessing higher concentrations of
 519 fumed silica.

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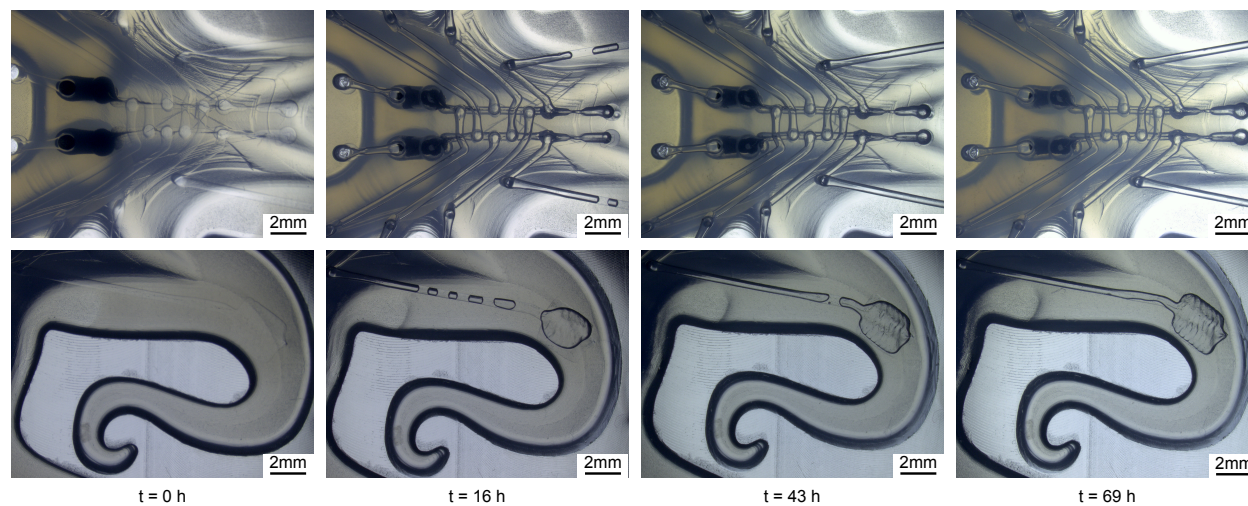


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 522 **Extended Data Figure 3. Modulus recovery of the body matrix after yielding.** **a**, A plot of
 523 storage modulus (G') as a function of time illustrates how the body matrix's modulus recovers
 524 during three-phase thixotropy tests. After a probe phase, a shear stress of 100 Pa is applied for
 525 varying times during a deformation phase, resulting in temporary fluidization of the matrix
 526 material. During the recovery phase, the modulus increases over time. **b**, $\tan(\delta)$, the ratio of the
 527 loss modulus (G'') to the storage modulus (G'), is plotted as a function time for each of the
 528 recovery phases measured in **a**. Onset of recovery of the body matrix material's yield stress –
 529 and the onset of fumed silica filler percolation in a recovering matrix material – is assumed to be
 530 the moment $G' = G''$, or $\tan(\delta) = 1$. Thus, the body matrix material's "recovery time" is
 531 approximately the time at which $\tan(\delta) = 1$ after deformation. Since the momentary deformation
 532 incurred by nozzle translation through a discrete volume of matrix material during EMB3D
 533 printing happens within a time period shorter than 1 s and with a magnitude less than 100 Pa, the
 534 body matrix material's thixotropic recovery time is less than 200 s, the approximate time it takes
 535 the body matrix to recover after sheared by a 100 Pa stress for 1 s.

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540 **Extended Data Figure 4. Rheological and printing behavior of inks and matrix materials**
541 **used to fabricate an Octobot.** **a**, A log-log plot of apparent viscosity as a function of shear rate
542 is provided for the fugitive ink (red), catalytic ink (black), body matrix material (blue), and fuel
543 reservoir matrix material (green). **b**, An octobot with fluorescently dyed fugitive inks (red, not
544 auto-evacuated) and hyperelastic actuator layers (blue) fabricated by molding and EMB3D
545 printing.

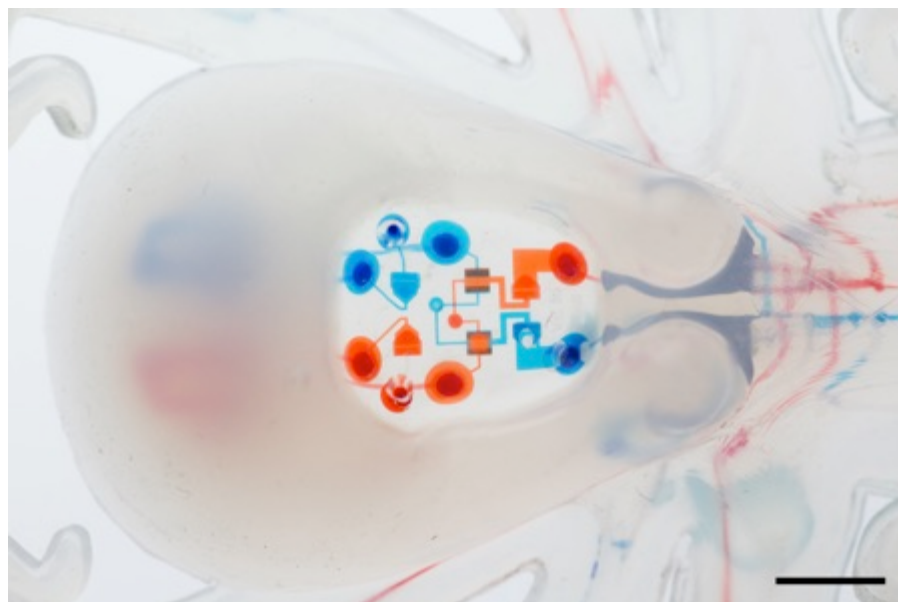
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Extended Data Figure 5. Auto-evacuation of the fugitive and catalytic inks. Photographs of an octobot's reaction chambers with upstream portions of the actuator networks (top) and a one-pad actuator at various times, t , reveal the auto-evacuation of the fugitive and catalytic inks, which leaves behind open channels that serve as mesofluidic features

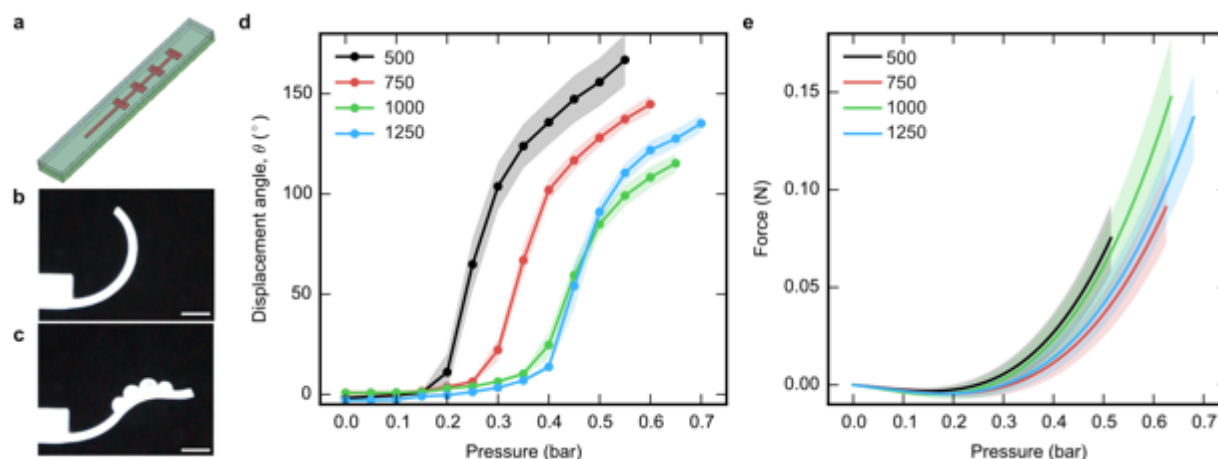
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Extended Data Figure 6. Infilling the soft controller from the fuel inlets. Water (with red or blue dye) is introduced into the fuel reservoir via the fuel inlets. Continuity between the fuel reservoirs, soft controller, and downstream EMB3D printed components is possible because of the fugitive plugs, which auto-evacuate along with the EMB3D printed inks. (The scale bar indicates a length of 5 mm.)

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 563 **Extended Data Figure 7. Characterization of EMB3D printed actuators.** **a**, CAD model of a
 564 four-bladder actuator design. Other bladder number, the design is similar to the actuators
 565 illustrated in Figure 4a. **b**, EMB3D printed actuator is shown prior to inflation. **c**, Actuator
 566 inflated to working pressure. (Scale bars in **b** and **c** indicate 5 mm.) **d**, Pressure versus
 567 displacement curves for four-bladder actuators with varying thickness of hyperelastic layer. **e**,
 568 Pressure versus force curves varying thickness of hyperelastic layer. (For **d** and **e**, thicknesses of
 569 the hyperelastic layer shown are in μm . Shaded regions indicate 95% confidence interval.)
 570 Detailed procedures for characterizing actuator performance are provided in the Methods.

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573 **Table S1. Break-in and working gauge pressures for EMB3D printed actuators.**

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Hyperelastic layer thickness (mm)	Two bladder actuators		Four bladder actuators	
	P_0 (Bar)	P_1 (Bar)	P_0 (Bar)	P_1 (Bar)
500	0.35	0.3	0.6	0.55
750	0.4	0.35	0.65	0.6
1000	0.45	0.4	0.7	0.65
1250	0.5	0.45	0.75	0.7
500	0.55	0.5	0.8	.075

575

575 List of Supplementary Videos and Tables

576 Supporting Video 1. EMB3D printing of an octobot.

577 Supporting Video 2. Decomposition of monopropellant fuel in the presence of catalyst.

578 Supporting Video 3. Octobot operation demo (front view).

579 Supporting Video 4. Octobot operation demo (top down view).

580

581 Table S1. Break in and working gauge pressures for EMB3D actuators.