



# 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.<sup>1,2</sup> Yet, despite recent advances, soft robots**  
19 **still remain tethered to hard robotic control systems and power sources.<sup>3-12</sup> 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 logic<sup>13</sup>**  
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.<sup>14</sup> 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.<sup>15,16</sup>**  
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,<sup>17,18</sup>  
36 electroactive polymers,<sup>19</sup> granular media,<sup>20</sup> and elastomers<sup>5,21</sup> exhibit elastic moduli ranging from  
37 100kPa to 1MPa,<sup>1</sup> are physically resilient,<sup>22,23</sup> and have the ability to passively adapt to their  
38 environment.<sup>1,2</sup> Molded and laminated elastomers with embedded pneumatic networks are  
39 widely used materials in soft robotics.<sup>1,21</sup> 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.<sup>1</sup> Robotic end effectors with bioinspired<sup>7</sup>  
42 and rapid<sup>24</sup> actuation, deployable crawlers<sup>3,10</sup> and swimmers<sup>11</sup> with complex body motions, and  
43 robust jumpers<sup>6,12</sup> 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.<sup>4,8,10-12</sup>

46 Creating a new class of fully soft, autonomous robots<sup>25</sup> 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.<sup>4,14</sup> 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 logic<sup>13</sup> 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<sup>13,26</sup> 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 behavior<sup>27</sup> (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.<sup>15,16</sup> 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.<sup>15,28</sup> 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<sup>15,16</sup> (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.<sup>15,28</sup> 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°C<sup>29</sup> (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,<sup>13</sup> 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).<sup>14</sup> 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 valves<sup>10</sup> 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  $\sim 6.4 \text{ mL}/\text{min}$ .<sup>13</sup> 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  $\mu\text{m}$ , 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  $\mu\text{m}$  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.<sup>13</sup> 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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167

**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.,<sup>26</sup> 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.<sup>15</sup> 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  $18 \text{ mW/cm}^2$  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  $4^\circ\text{C}$  in a syringe. When used, the fugitive plug material is allowed to physically gel  
233 before it is crosslinked for 3 min at  $6 \text{ mW/cm}^2$  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,  $2^\circ$  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 ( $10^{-2} \text{ s}^{-1}$ ) to high ( $10^2 \text{ s}^{-1}$ ) 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.<sup>30</sup> 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  $P_1$   
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  $P_1$ . 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  $P_1$ . 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.<sup>28</sup> 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  $6 \text{ mW/cm}^2$  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  $18 \text{ mW/cm}^2$  for 15 min to crosslink the catalytic ink. The mold is then transferred to a  $90^\circ\text{C}$   
315 oven, where the matrix materials crosslink. The octobot is removed from the mold and kept at  
316  $90^\circ\text{C}$  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  $\text{CO}_2$  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  $90^\circ\text{C}$  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

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406

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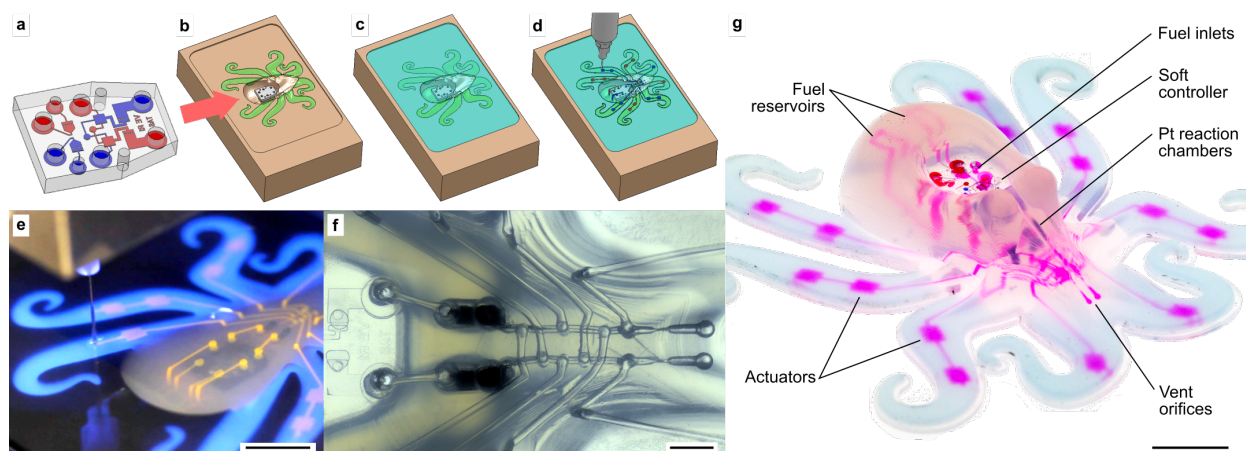
#### 417 **Author Contributions**

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**

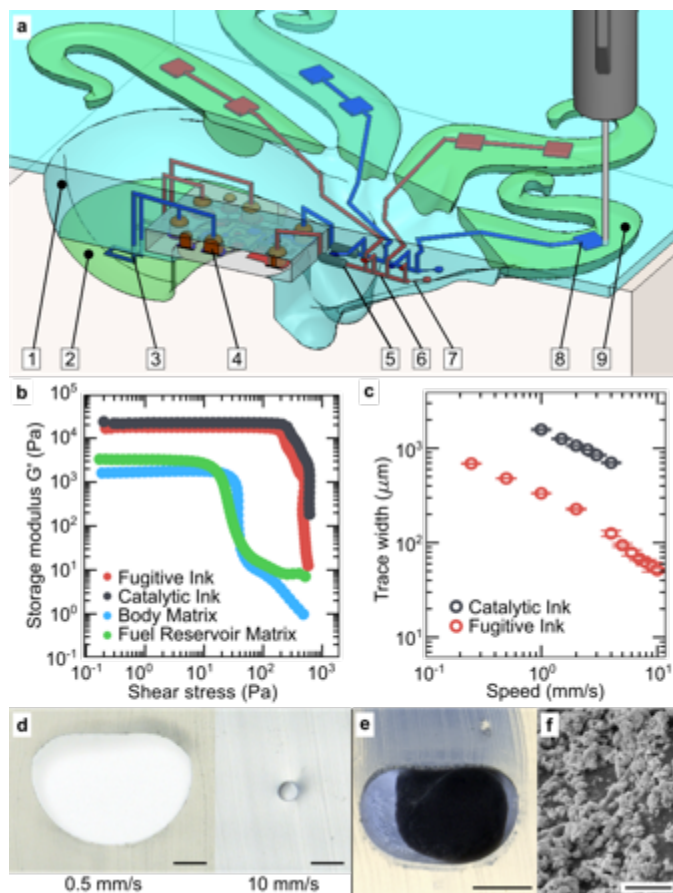
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423 declare no competing financial interests. Correspondence and requests for materials should be  
424 addressed to R.J.W. ([rjwood@seas.harvard.edu](mailto:rjwood@seas.harvard.edu)) and J.A.L. ([jalewis@seas.harvard.edu](mailto:jalewis@seas.harvard.edu)).

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426 **Figures and Figure Legends**

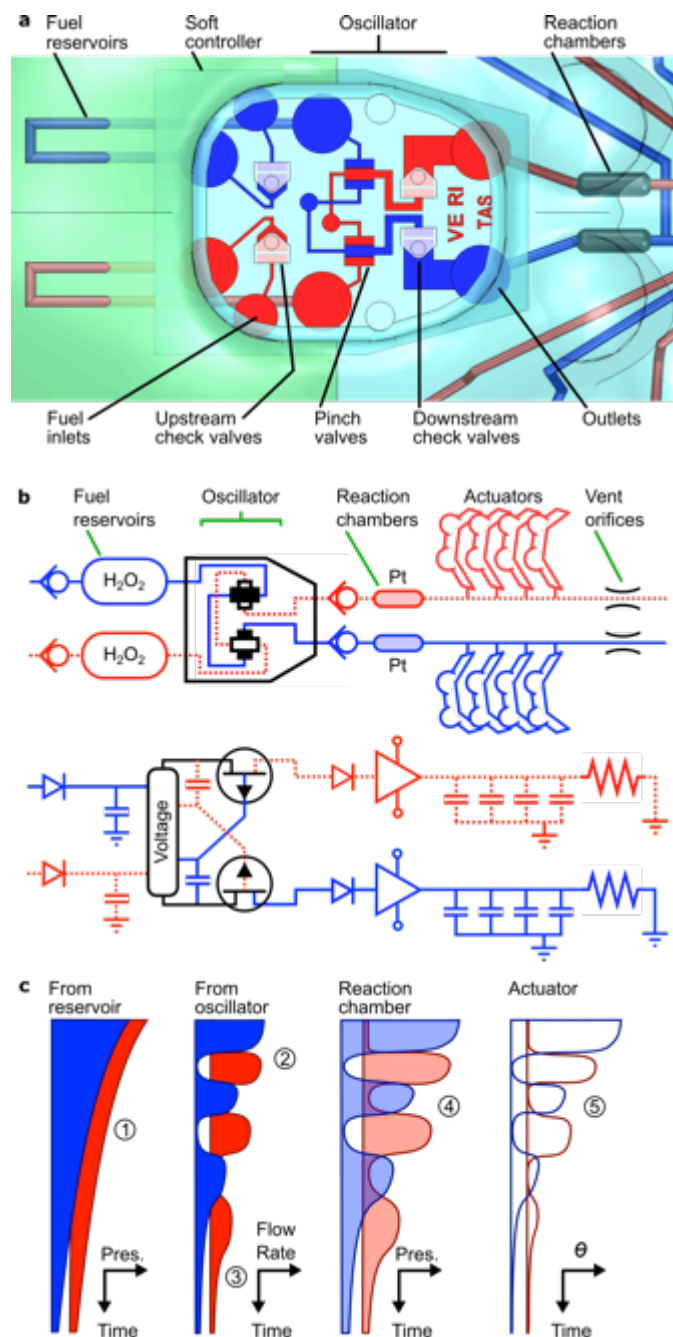
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 428 **Figure 1. Fully soft, autonomous robot assembly.** **a**, Pre-fabricate a microfluidic soft  
 429 soft controller, and **b**, load this controller into a mold. **c**, Pour matrix materials into the mold and **d,e**,  
 430 EMB3D print fugitive 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  $\mu\text{m}$ ).  
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  $\mu\text{m}$  and  
454 25  $\mu\text{m}$ , respectively.)



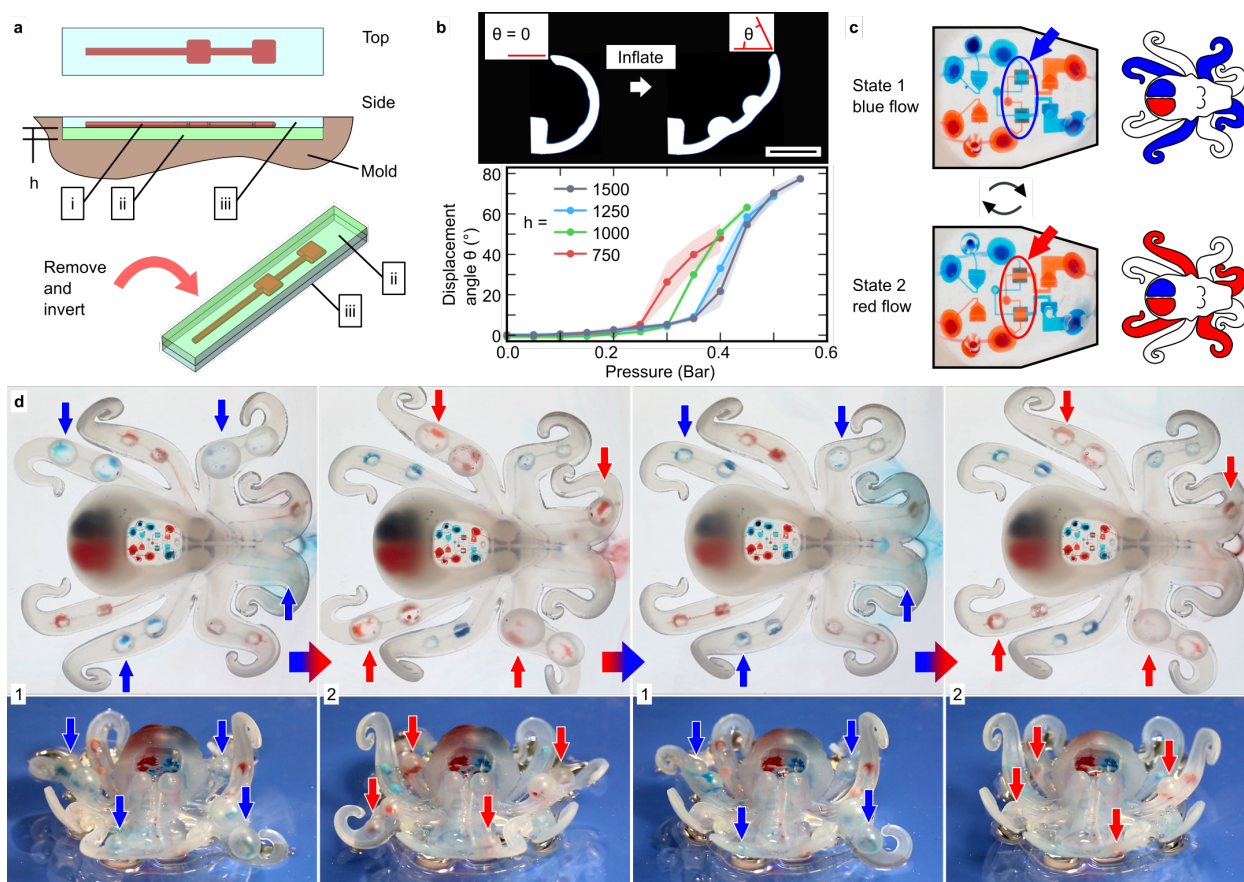
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**Figure 3. Octobot control logic.** Discrete sides are shown in red and blue for clarity. **a**, A system of check valves and switch valves within the soft controller regulates fluid flow into and through the system. **b**, A schematic and qualitative 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.

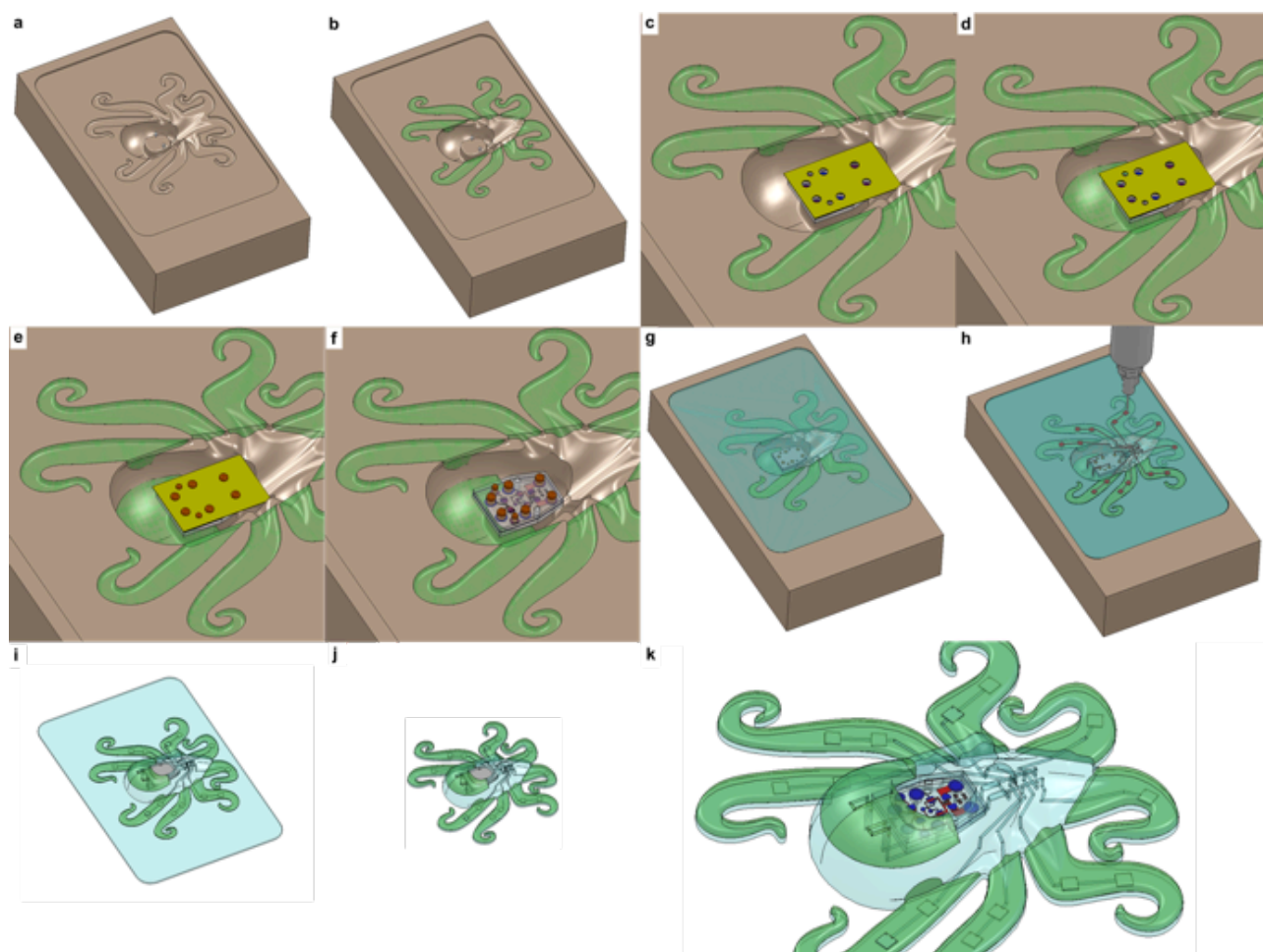


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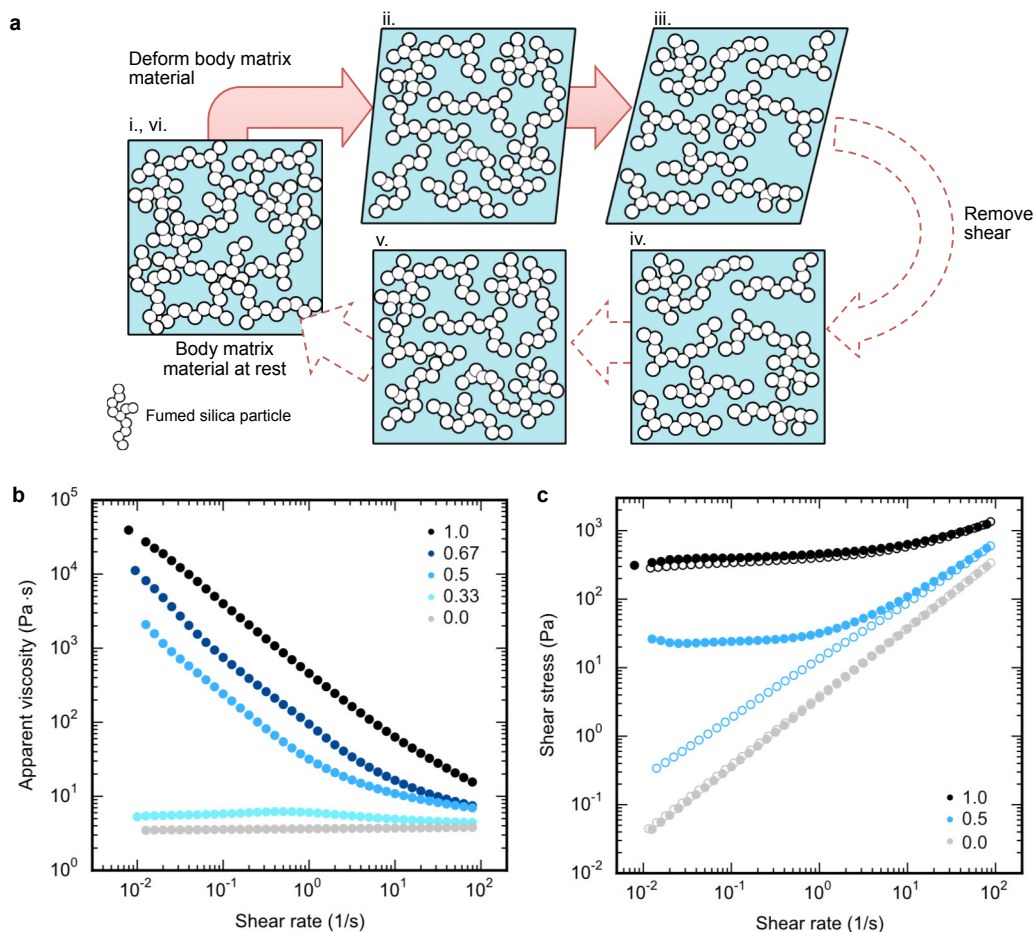
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 472 **Figure 4. Octobot actuation.** **a**, Actuator design in which traces (i) are printed in contact with  
 473 the hyperelastic layer (ii) inside of the body matrix material and (iii) differences in modulus  
 474 result in bending upon inflation. 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  $\mu\text{m}$ . **b**, (Top) The actuator tip angle  $\theta$  changes upon inflation (scale bar represents 10 mm).  
 477 (Bottom) Displacement angle  $\theta$  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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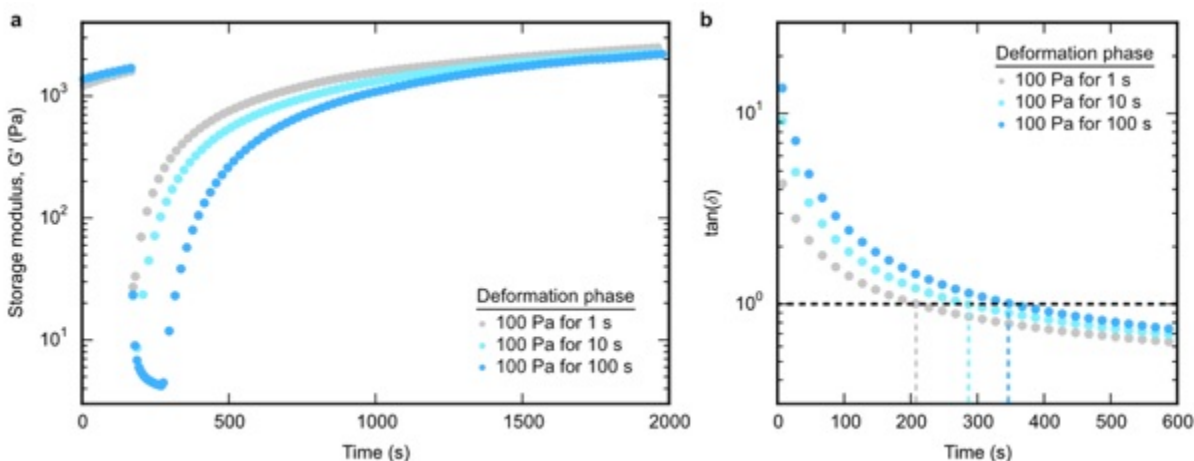
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485 **Extended Data Figure 1. Workflow for EMB3D printing an octobot.** **a**, An EMB3D printing  
 486 mold is machined from Delrin®. **b**, The hyperelastic layers needed for actuation are cast and  
 487 crosslinked in the mold's actuator regions. **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.

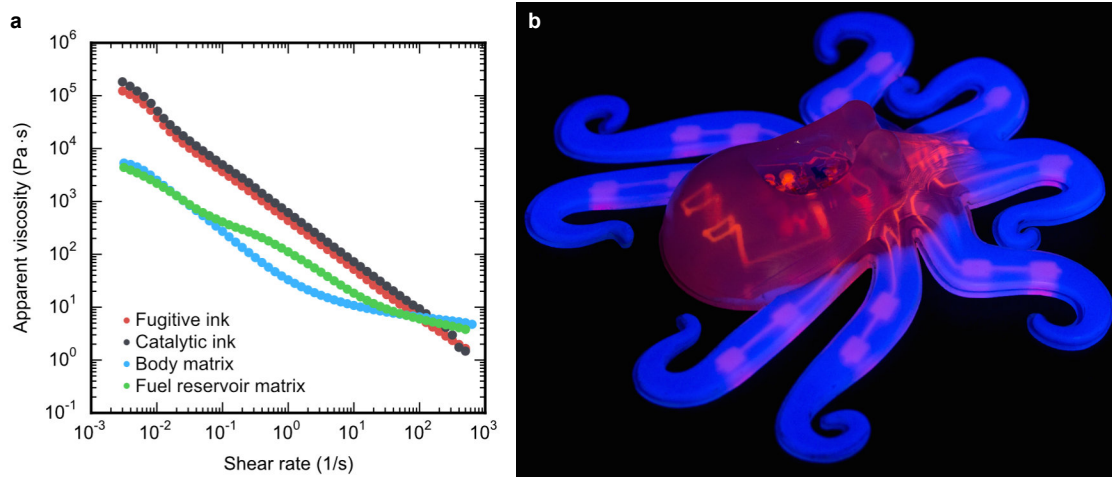


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 500 **Extended Data Figure 2. Rheological properties of the body matrix.** **a**, Schematic illustration  
 501 of the body matrix behavior during the EMB3D printing process. (i) When the body matrix is at  
 502 rest, the fumed silica fillers within 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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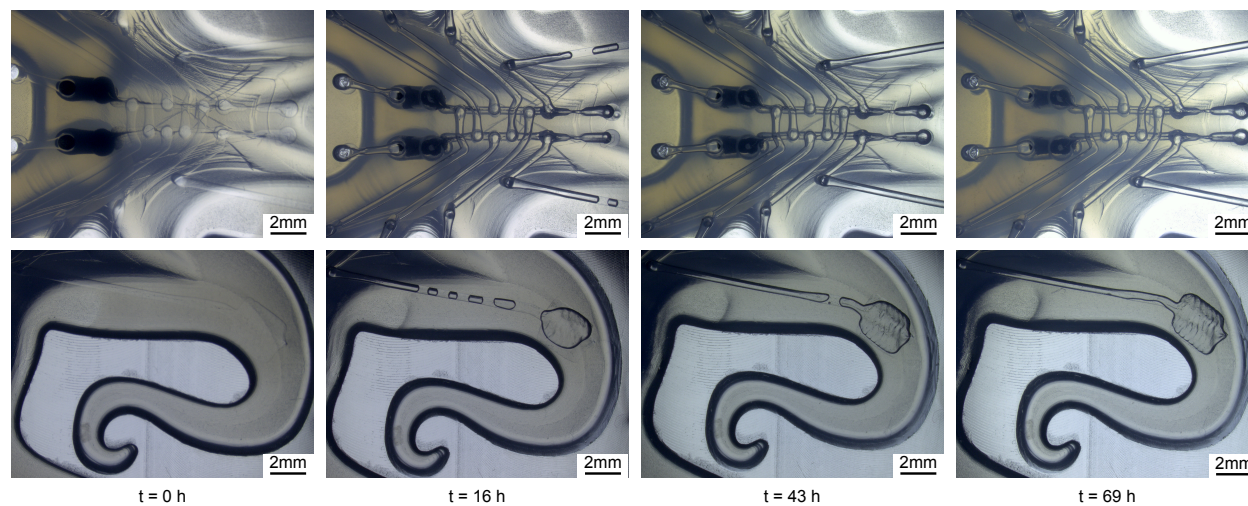


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 522 **Extended Data Figure 3. Modulus recovery of the body matrix after yielding.** **a**, A plot of  
 523 storage modulus ( $G'$ ) as a function of time illustrates how the body matrix's modulus recovers  
 524 during three-phase thixotropy tests. 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.

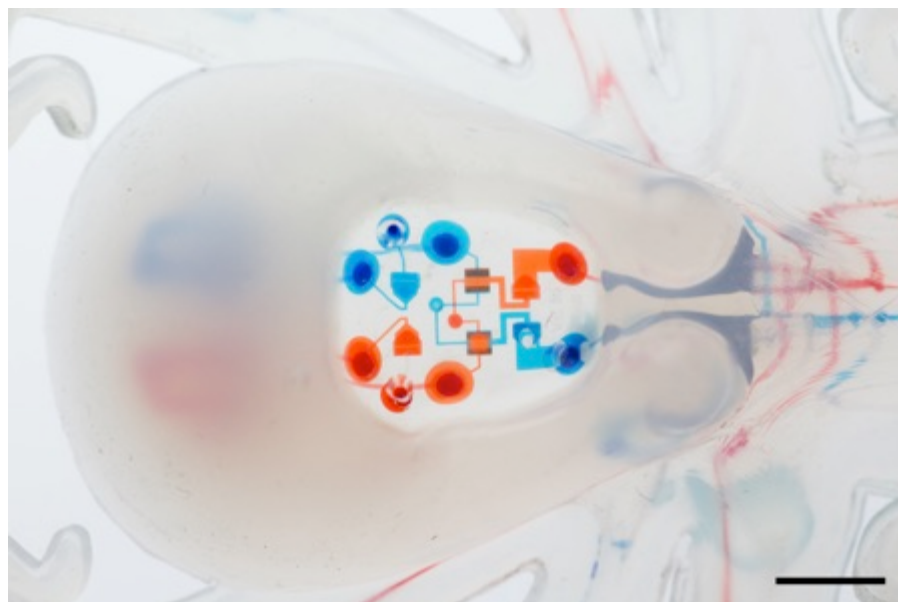
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**Extended Data Figure 5. Auto-evacuation of the fugitive and catalytic inks.** Photographs of an octobot's reaction chambers with upstream portions of the actuator networks (top) and a one-pad actuator at various times,  $t$ , reveal the auto-evacuation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

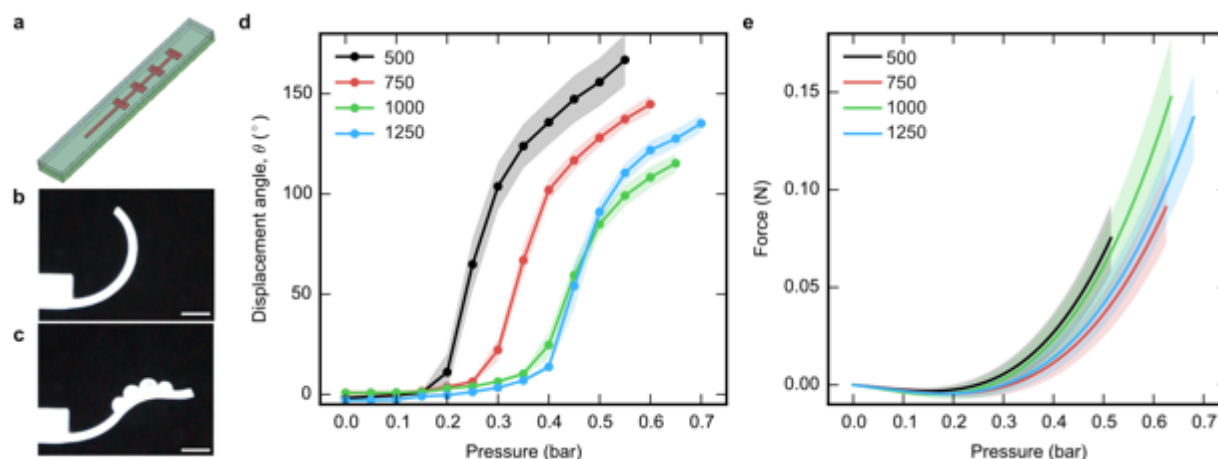
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**Extended Data Figure 6. Infilling the soft controller from the fuel inlets.** Water (with red or blue dye) is introduced into the fuel reservoir via the fuel inlets. Continuity between the fuel reservoirs, soft controller, and downstream EMB3D printed 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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564 **Extended Data Figure 7. Characterization of EMB3D printed actuators.** **a**, CAD model of a  
 565 four-bladder actuator design. Other bladder number, the design is similar to the actuators  
 566 illustrated in Figure 4a. **b**, EMB3D printed actuator is shown prior to inflation. **c**, Actuator  
 567 inflated to working pressure. (Scale bars in **b** and **c** indicate 5 mm.) **d**, Pressure versus  
 568 displacement curves for four-bladder actuators with varying thickness of hyperelastic layer. **e**,  
 569 Pressure versus force curves varying thickness of hyperelastic layer. (For **d** and **e**, thicknesses of  
 570 the hyperelastic layer shown are in  $\mu\text{m}$ . Shaded regions indicate 95% confidence interval.)  
 Detailed procedures for characterizing actuator performance are provided in the Methods.

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

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.