

Biodegradable electrohydraulic soft actuators

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September 14, 2022

Abstract

Biodegradable materials decompose and return to nature. This functionality can be applied to derive robotic systems that are environmentally friendly. This study presents a fully biodegradable soft actuator, which is one of the key elements in “green” soft robotics. The working of the actuator is based on an electrohydraulic principle, which is similar to that of hydraulically amplified self-healing electrostatic actuators. The actuator developed in this study consists of a dielectric film made of polylactic acid (PLA) and polybutylene adipate-co-terephthalate (PBAT), with soybean oil as the dielectric liquid and electrodes made from a mixture of gelatin, glycerol, and sodium chloride (NaCl). The synthesized biodegradable electrode material exhibits a Young’s modulus of 0.06 MPa and resistivity of 258 $\cdot$ m when the mass fraction of NaCl relative to the amount of gelatin and glycerol is 10 wt%. The softness and conductivity of the electrode material results in actuation strain values of 3.4% (at 1 kV, corresponding to 1.2 kV/mm) and 18.6% (at 10 kV corresponding to 9.6 kV/mm) for the linear-type and circular-type actuator, respectively. These values obtained for the biodegradable electrohydraulic soft actuators are comparable to those of non-biodegradable actuators of the same type, representing the successful implementation of the concept.

1. Introduction

Soft robotics has a high potential owing to the high compliance from which a wide variety of functional robots and applications can be derived.^[1–8] Synthetic polymers such as silicone rubbers are the most widespread materials used in soft robotics. They are low cost,^[9] easy to handle,^[10] and compatible with various fabrication methods, such as casting, molding, and printing.^[11] Synthetic polymers are also chemically stable, making them suitable for soft robots operated in diverse situations and environments, such as on the ground,^[12] underwater,^[13] in snowstorms,^[14] and even in radiation environments.^[15] On the contrary, their stable nature and irreversible synthetic process like thermoset^[16,17] make them non-biodegradable, which may lead to environmental destruction; this can particularly occur when the robots performing tasks in natural fields are discarded as the result of malfunctions or accidents. In addition, polymeric materials used in soft robotics are mostly difficult to recycle and have a high environmental impact. Considering these perspectives, it is important to incorporate biodegradability into soft robots.

Researchers have demonstrated biodegradable soft robotic elements that are focused on actuators. Their working principle includes pneumatic actuation,^[18–24] piezoelectricity,^[25] ion migration,^[26–30] and swelling.^[31,32] Pneumatic actuators are relatively easy to fabricate and can provide large outputs; however, their performance is dependent on bulky external pumps and compressors, which can lead to difficulty in constructing robots according to their types and specifications. From a system perspective, actuators based on piezoelectricity and ion migration have been driven electrically using a portable power source. However, actuation strain generated by piezoelectricity tends to be small (4%^[33]) and the actuation speed achieved with ion migration is normally low (2.3%/s^[34]), thus limiting the actuation performance. Similarly, actuation based on swelling

has a limitation on speed (over 6 h required for achieving a fully swelled state^[31]) and controllability of actuated deformation because its working principle requires material injection^[31] and cannot perform multiple actuations^[32].

In recent years, electrohydraulic soft actuators, also known as hydraulically amplified self-healing electrostatic (HASEL) actuators, are emerging.^[35] This type of actuators consists of a pair of opposing electrodes covering a portion of the surface of a flexible pouch encapsulating a dielectric liquid. When a high voltage is applied, electrostatic forces between the electrodes squeeze the pouch, causing the local position of the liquid to change, resulting in a hydraulic deformation of the entire structure as actuation. Electrohydraulic soft actuators exhibit large actuation strain (107% linear strain^[36]) and force (actuation stress of ~ 114 kPa^[36]), high power density (358 W/kg^[36]), and high speed (strain rate of 900%/s^[37]). Their structure is simple, allowing to tailor them in various shapes.

In this paper, we present a biodegradable soft actuator based on the electrohydraulic principle. This type of actuation principle requires compliant and conductive electrodes. First, we investigated the mechanical and electrical properties of the electrode for different compositions. Then, we fabricated and characterized two types of actuators that have linear and circular shapes to study the effect of incorporating biodegradable materials into the existing actuation principle and to validate our hypothesis.

2. Results and Discussion

The linear and circular biodegradable electrohydraulic soft actuators developed in this study are shown in **Figure 1a,b** (see also Supplementary Video S1). Both actuators have a sandwich structure composed of two layers of dielectric film made of polylactic acid (PLA) and polybutylene adipate-co-terephthalate (PBAT), forming a pouch encapsulating soybean oil as the dielectric liquid. PLA/PBAT film is known as a biodegradable substrate with good processability.^[38] The pouch is fabricated by welding two layers of dielectric film. On the pouch, electrodes are attached, which consist of a mixture of gelatin, glycerol, and sodium chloride (NaCl). The electrode material has an adhesive nature because of which it can be attached to the pouch without the need for glue. The entire surface of the device is covered by corn starch powder to keep the surface non-sticky. The integration of these materials and ingredients makes the actuator fully biodegradable. **Figure 2** shows a biodegradation process of the circular-type actuator. The device gradually degrades in the soil with time. The electrode material is not only biodegradable but also edible owing to the ingredients used. Therefore, the conductive material synthesized in this study may be useful in “edible robotics.”^[18] The geometry of our actuators is similar to those of the peano-HASEL actuator and circle planar HASEL actuator available in the literature.^[36,37] Since the dielectric liquid used is soybean oil, which provides no self-healing ability, we cease to use the abbreviation HASEL in the rest of the paper to avoid any confusion on the functionality.

The working principle of the linear and circular-type actuators is identical and is shown in **Figure 1c,d**. When voltage is applied, electrodes are charged, resulting in an electrostatic pressure. This pressure squeezes the pouch containing the soybean oil, which is placed in the areas where the electrodes do not overlap. The oil transferred in the non-electrode areas inflates the pouch, causing deformation of the entire structure. In the linear-type actuator, the actuated deformation occurs as contraction. In the circular-type actuator, it leads to expansion. In the linear-type actuator, the dimensions of the pouch and electrode area are 40 mm $\times$ 20 mm and 38 mm $\times$ 10 mm, respectively. In the circular-type actuator, the inner diameter of the pouch and the diameter of the electrode are 50 mm and 25 mm, respectively.

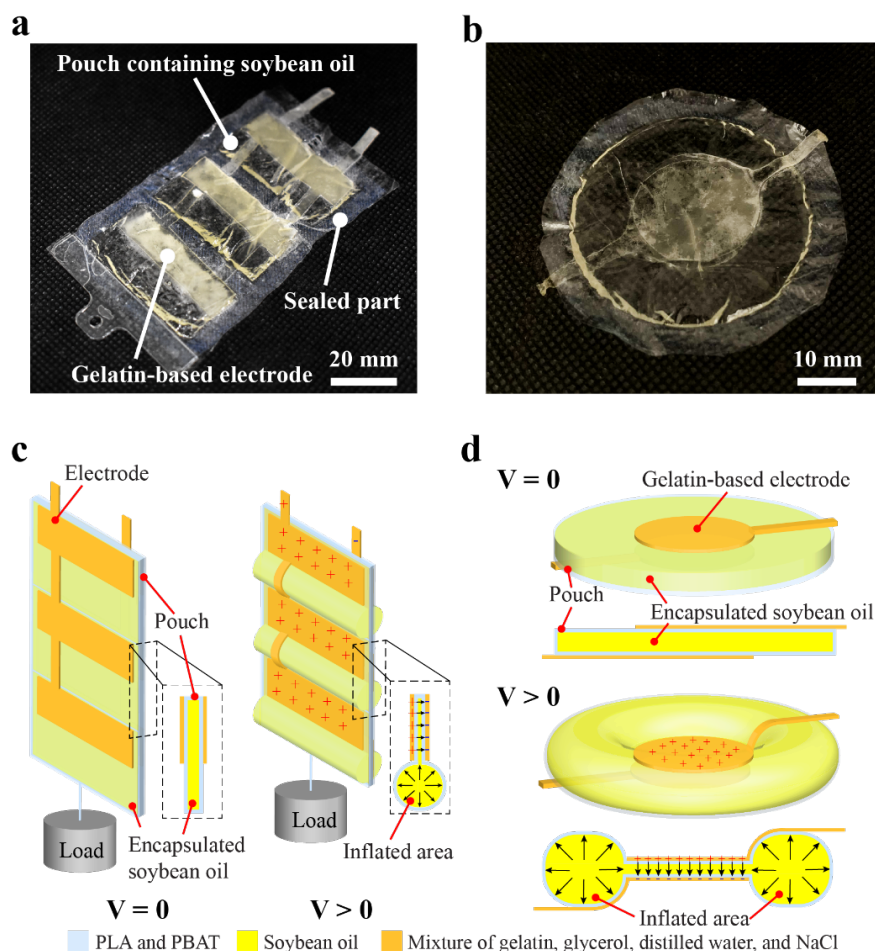


Figure 1. Biodegradable electrohydraulic soft actuators fabricated in this study. (a) Linear-type actuator. (b) Circular-type actuator. (c) Working principle of the linear-type actuator. (d) Working principle of the circular-type actuator.



Figure 2. Biodegradation process of the circular-type actuator. Right after fabrication, the actuator is placed on the soil, and the whole setup is contained in an oven to keep the environmental temperature constant to encourage microbial activity.

2.1. Electrical and mechanical properties of electrodes

We first assessed the electrical and mechanical properties of the electrode material to identify a composition suitable for the actuators. As mentioned previously, gelatin and glycerol were used as the base material of the electrodes. After curing, the mixture of gelatin and glycerol exhibited elastomer-like characteristics,

such as high stretchability (up to 209.6% of strain) and durability (multiple operations).^[18,21] Its electrical resistivity was $\sim 3500 \text{ } \Omega \cdot \text{m}$ according to our preliminary experiments, which limits the actuation of the biodegradable electrohydraulic soft actuators. Hence, we employed NaCl as an additive for the gelatin-glycerol mixture to enhance the conductivity and investigate its influence on the electrical and mechanical properties. Specifically, we measured the sheet resistance and Young's modulus of the electrode material for different amounts of NaCl (see Experimental Section for the details of the fabrication process of the electrodes and the measurements). The amounts of NaCl were 0, 1.25, 2.5, 3.75, and 5 g, which correspond to the mass fractions relative to the amount of gelatin and glycerol of 0, 2.7, 5.3, 7.7, and 10 wt%, respectively. Note that the mass fraction presented here does not include the presence of distilled water (initial mass 120 g) used in the mixing process of the electrode material. After curing, the water mostly evaporated and had an equilibrium state.

As shown in Figure 3a, the sheet resistance decreases with increasing fraction of NaCl. This indicates that the conductivity of the electrode material can be controlled by adjusting the amount of additive. For 10 wt%, the resistivity is $258 \text{ } \Omega \cdot \text{m}$, which is lesser than that of silicon ($640 \text{ } \Omega \cdot \text{m}$ ^[39]). This suggests that our material is conductive and can provide electric actuation in the devices developed in this study. Figure 3b shows the plot of the measured Young's modulus of the electrode material. The modulus takes the lowest value of 0.04 MPa for 0 wt% and increases up to 0.07 MPa until 5.3 wt%. Interestingly, from 5.3 wt% to 10 wt%, the modulus decreases from 0.07 MPa down to 0.06 MPa. The reason is as follows. Originally, gelatin binds to water molecules and forms a gel. Concurrently, sodium and chloride ions also bind to water molecules. This binding force is stronger in hydrated NaCl than in gelatin gel. Specifically, the binding force (van der Waals force) of gelatin molecules and water molecules is 0.4–4.0 kJ/mol.^[40] On the contrary, the ionic binding force is 20 kJ/mol.^[40] Therefore, hydrated NaCl binds the excess water in the sample (0–5.3 wt%) and hardens the electrodes, leading to a greater Young's modulus. However, from 5.3 wt% to 10 wt%, the water molecules originally bonded to gelatin now bond to NaCl, resulting in a lower Young's modulus.

The results show that there is a trade-off relationship between conductivity and compliance of the material. Depending on the application, softer and more conductive electrodes can be used, for instance, in soft electrically driven actuators. In this case, the presence of mechanically passive electrodes minimized the effect on actuation performance, and the electric current went through within a short time, thus ensuring a fast response. On the contrary, in some cases, it is difficult to pattern electrodes if they are too soft, making the fabrication complex.

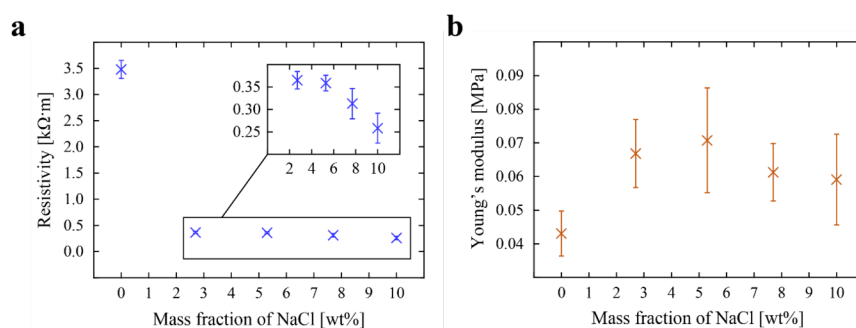


Figure 3. Characterization results of the gelatin-based electrodes. (a) Resistivity as a function of the NaCl mass fraction. (b) Young's modulus as a function of the NaCl mass fraction.

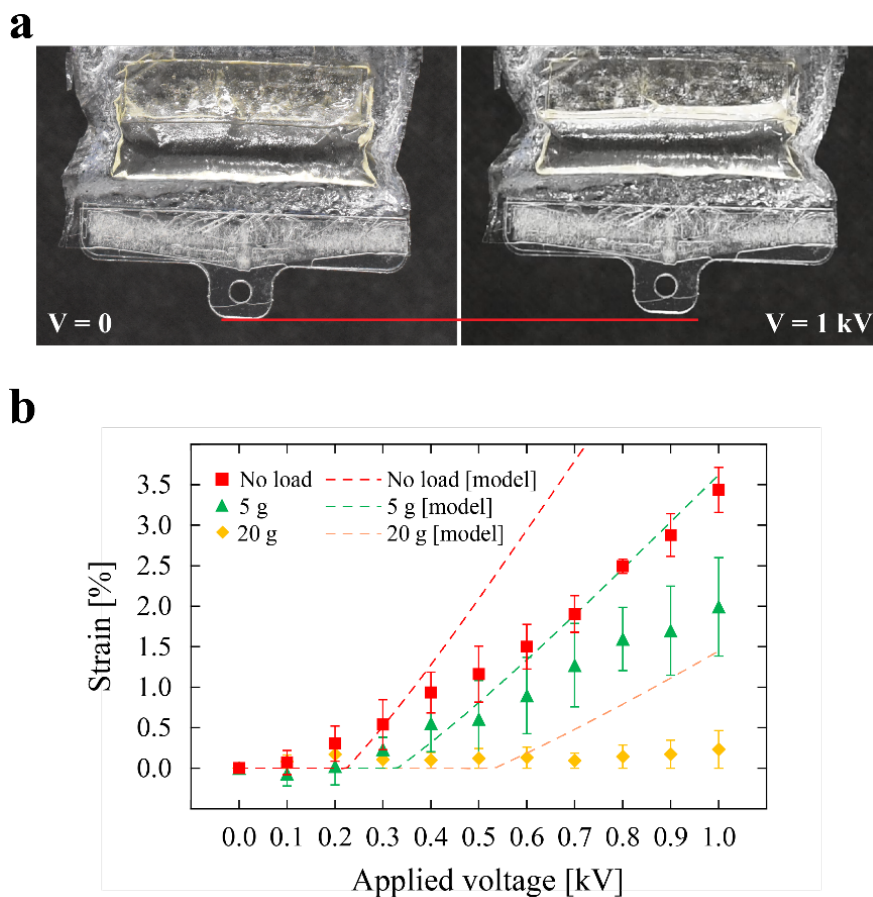


Figure 4. Characterization results of the linear type actuators. (a) Movement of the actuator. (b) Actuation strain as a function of the applied voltage.

2.2. Linear-type biodegradable electrohydraulic soft actuators

We performed an electromechanical characterization of the linear-type biodegradable electrohydraulic soft actuator. In this test, strain as a function of the applied voltage was measured under different loading conditions (0, 5, and 20 g). Electrodes with a NaCl mass fraction of 10 wt% were used in the tested actuators. The applied voltage was limited to 1 kV, as it was found during a preliminary experiment that the actuators undergo electrical breakdown at voltages more than 1 kV. This is reasonable given the thickness (10 μm) and the dielectric strength of the film material (in the case of PLA, $\sim 33 \text{ kV/mm}$ ^[41]). When actuated, the film layers of the pouch come closer owing to the electrostatic forces between the electrodes, resulting in the growth of the electric field strength. Figure 4 shows the actuation and characterization results of the actuators. The relationships for other tested actuators with different amounts of NaCl in their electrodes are also shown in Figure S6. As shown in Figure 4b, the actuation strain increases from 0% to 3.4%. Contrarily, the strain decreases under a larger applied load. The maximum strains in the tested voltage range for loading of 0, 5, and 20 g are 3.4%, 2.0%, and 1.4% respectively. The simulated values calculated by an analytical model (see Supporting Information for the details) exhibit the same trend as the experimental data, which suggests that the fabricated biodegradable actuators are functioning properly according to the fundamental working principle employed in this study. Thus, it is possible to design biodegradable electrohydraulic soft actuators with the aid of models intended to be used for non-degradable actuators. However, there is a discrepancy between the simulation and the experiment on the strain at each voltage. This may result from the fact that the simulation assumes a two-dimensional motion of the actuator, as shown in the cross-sectional

view of Figure 1c. The actuator also moves in three-dimensional motion, leading to actuation strains lower than the theoretical values. In addition, it also causes deformation of the sealed part of the pouch (5 mm width) that is mechanically passive.

At 1 kV, the measured strain of the biodegradable electrohydraulic soft actuators is 3.4%. This value is to that observed for non-biodegradable actuators of the same type; peano-HASEL exhibits an actuation strain of ~5.4% at 6 kV^[37]. The result indicates that biodegradable materials can be incorporated into electrically driven soft actuators without compromising the actuation strain.

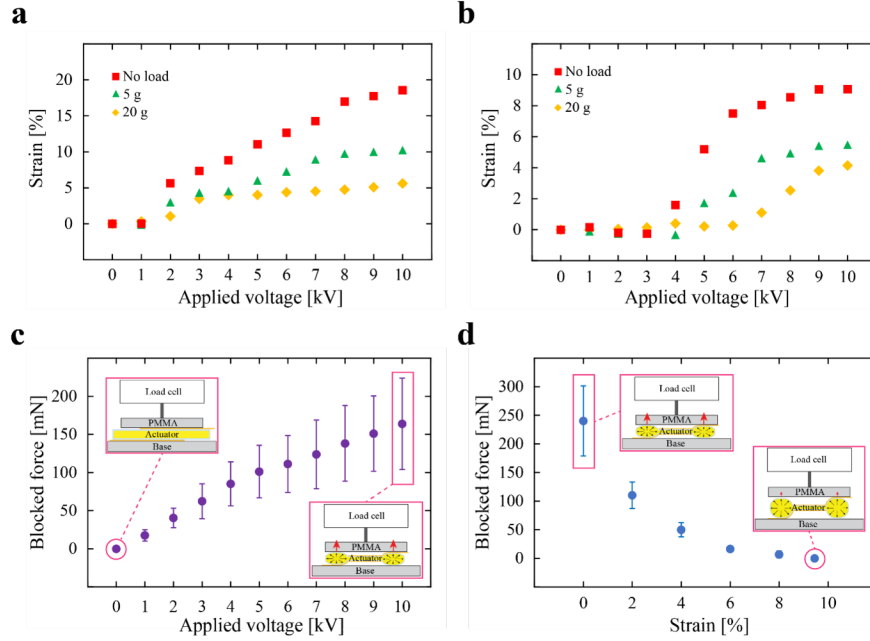


Figure 5. Characterization results of the circular-type actuators. (a) Actuation strain as a function of the applied voltage and electric field (amount of dielectric liquid 2 mL). (b) Actuation strain as a function of the applied voltage and electric field (amount of dielectric liquid 4 mL). (c) Blocked force as a function of the applied voltage and electric field. (d) Force-strain relationship at an applied voltage of 10 kV.

2.3. Circular-type biodegradable electrohydraulic soft actuators

Next, we characterized the circular-type actuators in terms of actuated strain and blocked force. The tested actuators have different amounts of soybean oil (dielectric liquid): 2 mL and 4 mL. The applied voltage ranges from 0 to 10 kV with a step size of 1 kV. Figure 5a,b display the measured strain as a function of the electric field under different loads (0, 5, and 20 g) for actuators with different amounts of soybean oil. Similar to the case of linear-type actuators, the actuation strain increases with the voltage and reaches 18.6% (2 mL soybean oil) and 9.1% (4 mL soybean oil) at 10 kV under no load. The loading results in reduced strain, similar to what is observed for the linear-type actuator. Unlike the linear-type actuators, the circular ones allow the application of voltage up to 10 kV. This is because, during the actuation, the electrodes are always distant, that is, the electric field strength does not exceed the breakdown strength of the pouch layers due to the relatively large amount of soybean oil against the entire volume of the actuator. The observed actuation strain in the biodegradable circular actuator is 18.6% at 10 kV, which is within the same range as a non-biodegradable actuator with identical dimensions (25% at 10 kV^[36]).

In the measured data (Figure 4a,b), a plateau and pull-in transition can be seen, which are described as unique characteristics of actuators of this type.^[36] Pull-in transition is described as a feature when the electrostatic force exceeds the threshold of the restoring force. In Figure 5a and Figure 5b, pull-in transition

can be seen at 2 kV and 4 kV, respectively. This behavior has also been reported in literature.^[36] In the actuator, the electrostatic force between the electrodes scales with the distance between the electrodes. Short distance results in larger electrostatic forces, leading to more actuation strain. The amount of dielectric liquid determines the distance between the electrodes and hence the actuation strain, which can be seen in the data shown in Figure 5a,b, where different amounts of liquid (2 mL and 4 mL) are employed.

Next, we investigated the blocked force as a function of the applied voltage. As plotted in Figure 5c, the force increases with the voltage and reaches a value of 241 mN at 10 kV, corresponding to an actuation pressure of 0.12 kPa. We further examined the blocked force at different actuation strains, from which the force-strain characteristics of the actuators were assessed at 10 kV. As shown in Figure 5c, the blocked force reduces as the strain increases, which is also observed in non-biodegradable actuators of the same type^[42]. Further, to compare the actuation pressure, the non-biodegradable circular-type actuator exhibits a value of 25 kPa at 8 kV^[42]. This value is larger than that observed for our actuator, which could be attributed to the difference in the material properties of the dielectric liquids. For instance, a larger dielectric constant leads to a larger electrostatic force and hence a higher pressure. This implies that the output of biodegradable electrohydraulic soft actuators can be increased using dielectric liquids with a high dielectric constant. Nevertheless, the discussed results confirm that the actuators developed in this study function according to the working principle and their actuation performances are comparable to those of the non-biodegradable ones; this proves the successful implementation of our hypothesis.

3. Conclusion

In this study, we presented biodegradable electrohydraulic soft actuators as electrically driven green robotic elements for environmentally friendly soft robotics. We showed that the electrodes used for the actuators can be tuned to achieve both softness and conductivity suitable for electrically driven soft devices. We also demonstrated the actuators in different forms; both exhibited actuation performance comparable to non-biodegradable counterparts.

In the future, we will focus on the improvement of actuation performance by investigating suitable biodegradable materials for every part of the actuator, where both analytical and computational models are useful. Since the actuator presented in this study consists of dielectric and conductive materials, that is, essential components for electrically driven soft robotic elements, applying the materials to existing working principles may result in biodegradable soft robotic devices of various forms, such as stretchable sensors, electroadhesive pads, and soft pumps. Furthermore, the electrode fabricated in this study is essentially edible, and it can be applied to realize a broad range of devices in edible robotics.

Experimental Section

Materials

A dielectric film made of polylactic acid (PLA) and polybutylene adipate-co-terephthalate (PBAT) was purchased from Earth Friendly (“[Biodegradable 100%] Wrap returned to soil”). Gelatin powder (17009-01) and glycerol (17029-00) were purchased from Kanto Chemical. Sodium chloride (NaCl) was purchased from Tomita Pharmaceutical (7647-14-5). Corn starch powder and soybean oil were purchased from a supplier.

Mechanical characterization of electrode material

Gelatin powder was soaked in distilled water and left at room temperature (23) for 10 min. The amount of gelatin powder and distilled water were 30 g and 120 g, respectively. Then, glycerol and NaCl were added. The amount of glycerol was 15 g. The amount of NaCl was 0, 1.25, 2.5, 3.75, and 5 g, which corresponded to the mass fraction relative to the amount of gelatin and glycerol of 0, 2.7, 5.3, 7.7, and 10 wt%, respectively. The mixed materials were dissolved at 80°C and stirred at 200 rpm for 1 h using a hot stirrer (AS ONE, CHPS-170DF). The solution was poured into a mold and cured at room temperature for 1 h. The cured sample was demolded and placed in a humidity chamber (Tolihan, WET-297-AHU) at 23°C (67% RH) for at least 24 h until testing. The diameter and thickness of the samples were 29 mm and 12.5 mm, respectively. Three samples were used for each material composition (mass fraction of NaCl 0, 2.7, 5.3, 7.7, and 10 wt%).

The testing procedure followed was ISO 7743. Each sample was compressed at a speed of 10 mm/min using a universal testing machine (Shimadzu, AGS-20NX) from which the stress-strain curve was obtained. The testing machine performed four reciprocating motions compressing from 0% to 25% of strain and rebounding from 25% to 0%. The measured force under 20% compressive strain of the fourth round was used to determine the Young's modulus. It was calculated by dividing the force by the cross-sectional area and strain of the specimen before compression.

Electrical characterization of electrode material

Gelatin powder was soaked in distilled water and left at room temperature for 10 min. The amounts of gelatin powder and distilled water are 30 g and 120 g, respectively. Then, glycerol and NaCl were added. The amount of glycerol was 15 g. The amount of NaCl was 0, 1.25, 2.5, 3.75, and 5 g, which corresponded to the mass fraction relative to the amount of gelatin and glycerol of 0, 2.7, 5.3, 7.7, and 10 wt%, respectively. The mixed materials were dissolved at 80 °C and stirred at 200 rpm for 1 h using a hot stirrer (AS ONE, CHPS-170DF). The solution was then blade-casted on a plastic substrate using a film applicator (Industrial Physics, TQC Sheen) and an applicator coater (Zehntner, ZUA 2000), forming an electrode layer with a thickness of 143 μm after curing at room temperature for 24 h at 23 °C (67% RH). The thickness of the layer was measured using a laser displacement sensor (OPTEx FA, CDX-L15). Four samples were prepared for each material composition (mass fraction of NaCl was 0, 2.7, 5.3, 7.7, and 10 wt%). The dimensions of the samples were 10 mm in length and 10 mm in width. The electrical resistance of every specimen was measured using a digital multimeter (Keithley, 2100). The resistivity was then calculated based on the measured resistance by considering the dimensions of the sample.

Fabrication of biodegradable electrohydraulic soft actuators

The fabrication process is illustrated in Figure S1 and Figure S2. For the linear-type actuators, two layers of biodegradable dielectric film were bonded by thermobonding. This was done using a 3D printer (Flashforge 3D technology, Adventurer 3). The print head nozzle of the 3D printer was heated to 180°C, and the platform was kept at 50 °C. By moving the head nozzle while keeping in contact with the dielectric film layers, a pouch of the designed dimensions was fabricated. Then, soybean oil was injected into the pouch. The amount of oil used was 1.4 mL. The gelatin-based electrode was prepared using the same steps as used in the fabrication of samples for the resistivity measurement. The gelatin solution heated at 80°C for 1 h was blade-casted on a plastic substrate using a film applicator (Industrial Physics, TQC Sheen) and an applicator coater (Zehntner, ZUA 2000), forming an electrode layer with a thickness of 143 μm after curing at room temperature for 24 h at 23°C (67% RH). Then, the casted electrode layer was cut using a plotter cutter (Graphtec, CE6000-40 Plus). The electrode was attached to the pouch made of biodegradable layers and corn starch powder was coated on it. The fabrication process of the circular-type actuators was the same as that of the linear-type, except for the amount of oil used (2 ml and 4 ml) and the thickness of the electrode layer (50 μm).

Characterization of linear-type actuators

The experimental setup is shown in Figure S3. The characterization was conducted by applying a voltage of up to 1000 V with 100 V incremental to the actuator. The voltage was supplied from a high-voltage DC-DC converter (XP Power, CB101). The actuation strain as a function of the applied voltage was then acquired by measuring the displacement of the actuator tip, which was done by using a camera (Nikon, D3500) followed by image processing.

Characterization of circular-type actuators

The characterization process is depicted in Figure S4. The actuation strain of the actuator was measured using a laser displacement sensor (OPTEx FA, CDX-150). Up to 10 kV of voltage was applied to the actuator with 1 kV incremental. A PTFE sheet was placed on the actuator so that the laser sensor can detect the actuated displacement accurately. For characterizing the strain-voltage relationship, the actuation force was measured at a certain strain using a linear stage (Thorlabs, PT 1/M). The force generated by actuation was measured using a load cell (FUTEK, LSB 200). The load cell was placed above the actuator to identify the

accurate normal force. The initial load was applied, the mass of which was identical to the PTFE sheet.

Degradation test of the actuator

The circular-type actuator was placed on actinomycetes (Oki, Dr. Actinomycete)-mixed soil in an oven (Yamato Scientific, ADP 300C) at 60 °C. The oven had a small hole that allowed air to flow in, which kept the actinomycete active. The soil was collected on the campus of the University of Electro-Communications.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgments

This work was supported by the JSPS KAKENHI Grant-in-Aid for Scientific Research on Innovative Areas “Science of Soft Robot” project (grant number 21H00324).

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Received: ((will be filled in by the editorial staff)) Revised: ((will be filled in by the editorial staff)) Published online: ((will be filled in by the editorial staff))

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Biodegradable electrohydraulic soft actuators

R. Kanno, F. Caruso, K. Takai, Y. Piskarev, V. Cacucciolo, D. Floreano, J. Shintake*

Fully biodegradable soft actuators based on the electrohydraulic principle are presented. The actuators exhibit performance comparable to that of non-biodegradable actuators. This work is expected to contribute to the realization of electrically driven soft robots that are environmentally friendly.

Keywords: soft robotics, green robotics, biodegradable, edible electrode, electrohydraulic soft actuators, hydraulically amplified self-healing electrostatic actuators



Supporting Information

Biodegradable electrohydraulic soft actuators

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This section presents a model to describe the voltage-strain response of biodegradable Peano-HASEL actuators with different external loads F applied. The model is based on work in the literature for non-biodegradable HASELs [1,2]. A detailed description of the working principle of these systems is given in the result and discussion section. Here, we describe the mathematical model and all the assumptions.

Geometric assumptions The model considers only in-plane deformations and is based on the assumption that the shape of the liquid pouch, in each configuration, can be approximated by two symmetric circular arcs, as shown in Figure 1. With this assumption, the actuator stroke x is fully described by the angle at the zipping point α

$$x(\alpha) = h - \left(l_e(\alpha) + l_p(\alpha) \frac{\sin(\alpha)}{\alpha} \right) \quad (1)$$

where h is the actuator height at rest, $l_e(\alpha)$ and $l_p(\alpha)$ are the length of the zipped and unzipped portions of the pouch, respectively. Since the liquid volume is constant during deformation, so is the area A in each 2D cross section. $l_e(\alpha)$ and $l_p(\alpha)$ are a function of the area A and of the angle α

$$A = A_{\text{def}} = \frac{1}{2} L_p^2 \left(\frac{\alpha_0 - \sin(\alpha_0) \cos(\alpha_0)}{\alpha_0^2} \right) \quad (2)$$

$$l_p(\alpha) = \sqrt{\frac{2A\alpha^2}{\alpha - \sin(\alpha) \cos(\alpha)}} \quad (3)$$

$$A = A_{\text{def}} = \frac{1}{2} L_p^2 \left(\frac{\alpha_0 - \sin(\alpha_0) \cos(\alpha_0)}{\alpha_0^2} \right) \quad (2)$$

$$l_e(\alpha) = L_p - l_p(\alpha) \quad (4)$$

where L_p is the length of the pouch at rest.

Electrostatic assumptions

We assume that the change in energy of the actuator is only due to the increased capacitance due to the increased length of the zipped region $l_e(\alpha)$. We neglect the change in the electric field in the unzipped region (where the dielectric fluid is) when changing the angle α . We assume ideal dielectrics. We assume there is no liquid in-between the electrodes in the zipped region (the two electrodes are only separated by the dielectric film). As a result, the capacitance of the actuator as a function of α is

$$C(\alpha) = \frac{\varepsilon_r \varepsilon_o w}{2t} l_e(\alpha) \quad (5)$$

where w is the width of the actuator, t the thickness of the dielectric film, ε_r and ε_o the relative and vacuum permittivities, respectively.

Total Free energy of the system

The Helmholtz free energy of the system can be expressed as

$$U_t(\alpha, V) = U_e(\alpha, V) + U_m(\alpha) - qV \quad (6)$$

where $U_e(\alpha, V) = \frac{1}{2} C(\alpha) V^2$ is the electrostatic energy stored in the capacitor, $U_m(\alpha) = Fx(\alpha)$ is the mechanical work done by the external load F , and $-qV$ is the electrical work supplied by the battery. We neglect the bending strain energy and gravitational energy of the dielectric film given its low thickness and relatively large external loads.

Equation 6 can be modified (using the relationship $q = CV$)

$$U_t(\alpha, V) = Fx(\alpha) - \frac{1}{2} C(\alpha) V^2 \quad (7)$$

Force-load relationship

The relationship between the external load F and the angle α , at each voltage value V obtained by minimizing Eq.(7), as $dU_t(\alpha, V)/d\alpha = 0$ and solving for F

$$F = \frac{w \cos(\alpha) \varepsilon_r \varepsilon_o V^2}{4t (1 - \cos(\alpha))} \quad (8)$$

The strain of the actuator is obtained as

$$\epsilon(\alpha) = \frac{x(\alpha)}{h} = 1 - \frac{\alpha_0}{\sin(\alpha_0)} \left(1 + \frac{\sqrt{2A}}{L_p} \frac{\sin(\alpha) - \alpha}{\sqrt{\alpha - \sin(\alpha) \cos(\alpha)}} \right) \quad (9)$$

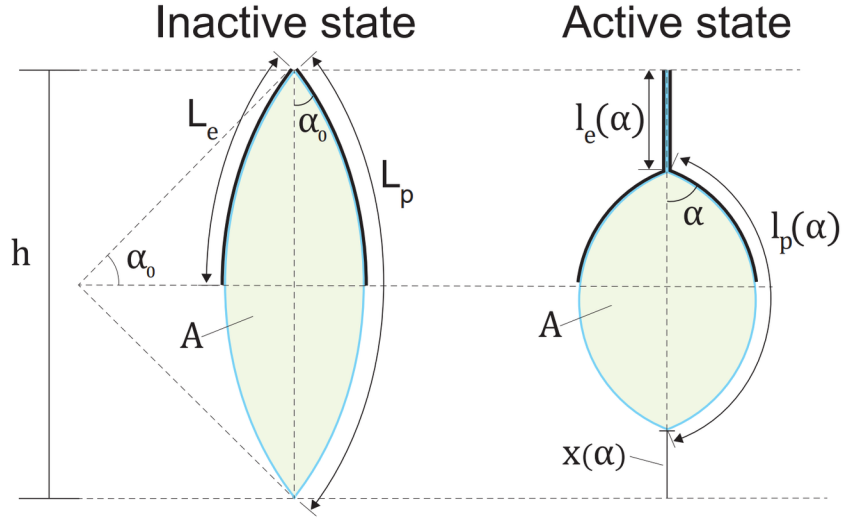
Combining equations (8 and 9) we obtain the voltage-strain relationship for a single Peano-HASEL actuator.

$$\epsilon(a) = \frac{\alpha_0}{\sin(\alpha_0)} \left(\frac{\sqrt{2A} \left(\arccos\left(\frac{4Ft}{c_2}\right) - c_1 \right)}{L_p \sqrt{\arccos\left(\frac{4Ft}{c_2}\right) - \frac{4Ftc_1}{c_2}}} - 1 \right) + 1 \quad (10)$$

where

$$c_1 = \sqrt{1 - \frac{16F^2t^2}{c_2^2}} \quad (11)$$

$$c_2 = \epsilon_0 \epsilon_r w V^2 + 4Ft \quad (12)$$



Geometry of a Peano-Hasel actuator.

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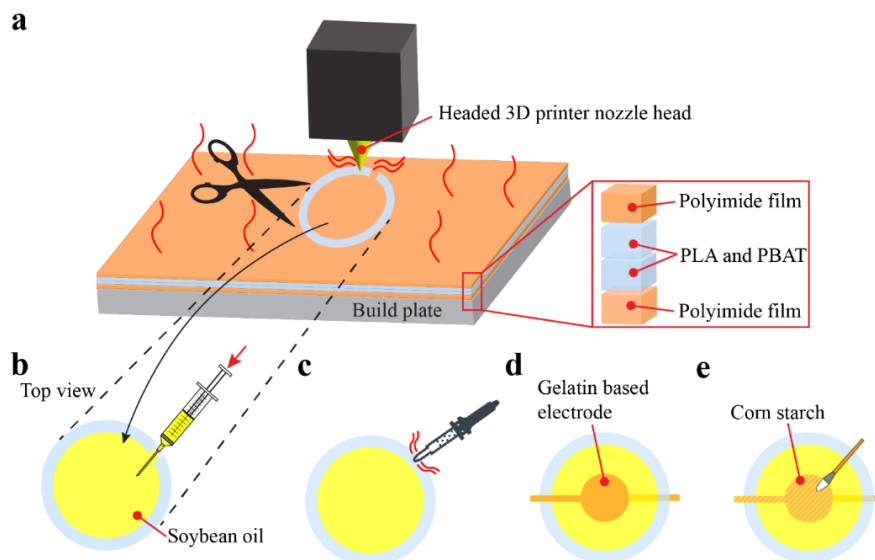


Figure S1. Fabrication procedure of circular-type actuators. (a) Sandwiched between dielectric polyimide films and welded by a 3D printer. (b) Soybean oil inserted into the welded dielectric film using a syringe. (c) Welded by soldering iron. (d) Sandwiched with electrodes. (e) Corn starch coated on the electrodes.

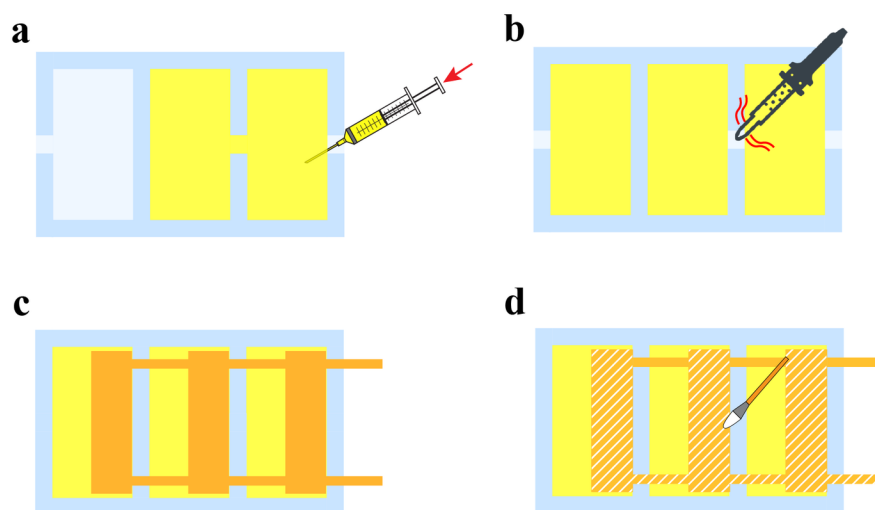


Figure S2. Fabrication procedure of linear-type actuators. Before processing from (a) to (d), two sheets of dielectric film were welded by a 3D printer. (a-b) Soybean oil is injected into the dielectric film using a syringe. (c) Electrodes are attached. (d) Corn starch is coated on the electrodes.

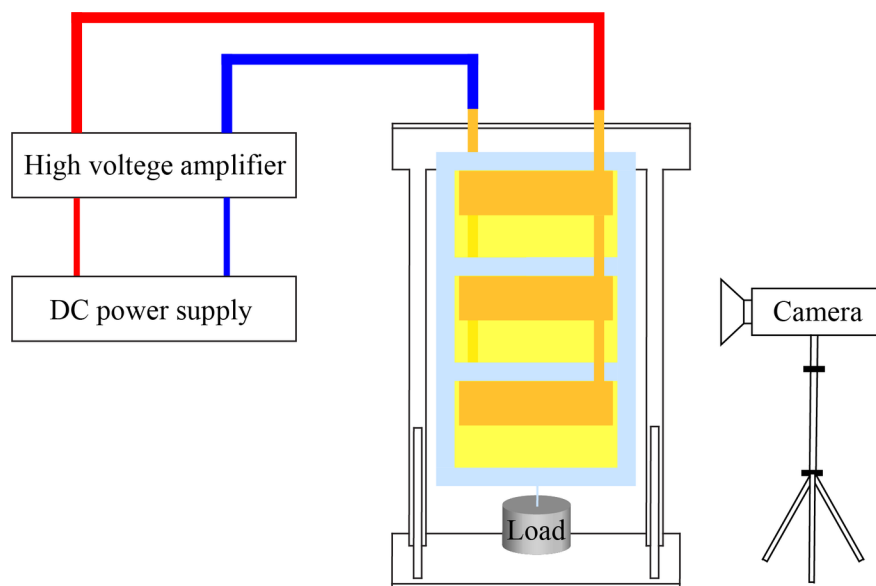


Figure S3. Measurement setup for the linear-type actuators. The actuator is actuated by a high voltage amplifier. DC power supply provides a constant voltage. The actuation strain was measured by a camera.

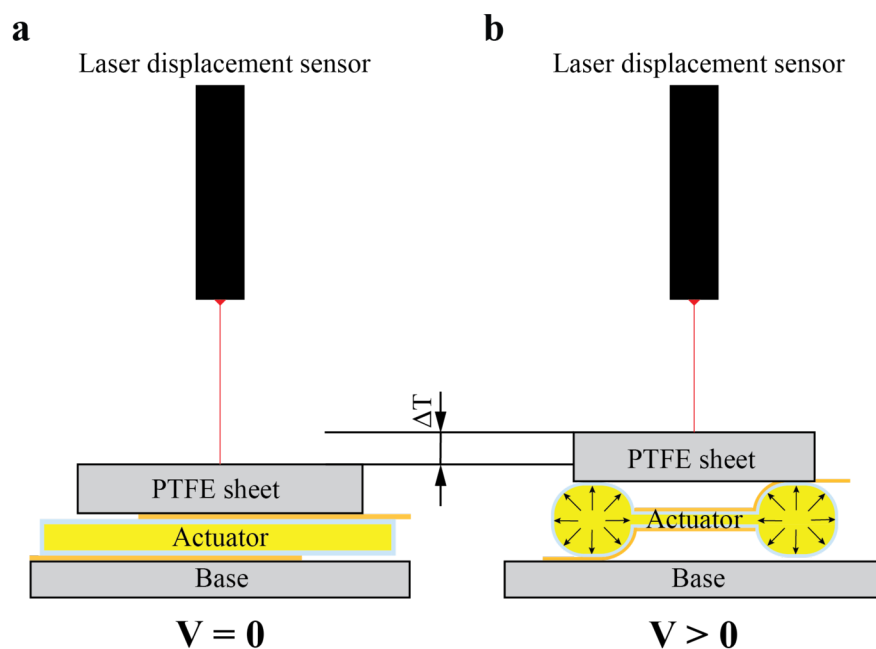


Figure S4. Strain measurement setup of circular-type actuators. PTFE sheet (8.8 g) was placed on the actuator, which is used as the initial load.

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Figure S5. Blocked force versus strain characterization setup of circular-type actuators. (a) Initial load (8.8 g: same as the weight of the PTFE sheet used in strain measurement setup) is applied to the actuator without an applied voltage. (b) 10 kV is applied by fixing the position of the load cell and the blocked force is measured. (c) Load cell position is elevated and the blocked force is measured by maintaining a 10 kV voltage supply.

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Figure S6. (a) Actuation strain as a function of the applied voltage (2.7 wt% electrodes).
(b) Actuation strain as a function of the applied voltage (5.3 wt% electrodes).