

# Realizing the potential of dielectric elastomer artificial muscles

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**Soft robotics represents a new set of technologies aimed at operating in natural environments, and near the human body. To interact with their environment, soft robots require artificial muscles to actuate movement. These artificial muscles need to be as strong, fast, and robust as their natural counterparts. Dielectric elastomer actuators (DEAs) are promising soft transducers, but typically exhibit low output forces and low energy densities, when used without rigid supports. Here we report a soft composite DEA made of strain stiffening elastomers and carbon nanotube electrodes, which demonstrates a peak energy density of 19.8 J/kg. The result is close to the upper limit for natural muscle (0.4-40 J/kg) making these DEAs the highest performance electrically driven soft artificial muscles demonstrated to date. To obtain high forces and displacements, we used low density, ultra-thin carbon nanotube electrodes which can sustain applied electric fields upwards of 100 V/ $\mu$ m without suffering from dielectric breakdown. Potential applications include prosthetics, surgical robots, and wearable devices, as well as soft robots capable of locomotion and manipulation in natural or human-centric environments.**

artificial muscles | soft robotics | dielectric elastomer actuators

Soft robotics is a nascent sub-field of robotics, with a broad range of applications(1–3), from wearable and rehabilitative devices(4), to novel surgical tools(5), as well as biomimetic robots for search, rescue, and exploration(6, 7). A fundamental challenge in developing these robots is the creation of effective actuators to deform a soft body to achieve locomotion, manipulation, or other tasks(8, 9). The gold standard for actuation in a soft body is natural muscle(10), which exhibit high energy density (0.4-40 J/kg), a broad range of frequencies (1-200 Hz), and large strains (5-30%). Since their initial development, dielectric elastomer actuators(11–13) (DEAs) have been hailed as artificial muscles, but material limitations typically constrain the user to choose between a truly soft actuator which exhibits low energy density(14) and an energy dense actuator with multiple rigid components(15). Fundamentally, DEAs operate as compliant capacitors, in which the dielectric elastomer deforms in response to an applied electric field. In this paper we describe how with careful material selection and processing, a DEA can be made to operate as a strong artificial muscle without requiring any rigid components.

In this article we realize a long standing vision for DEAs as fully soft, electrically driven, energy dense artificial muscles (Fig. 1 A,B). Single film dielectric elastomers are usually thin, to minimize the actuation voltage, which leads to small force outputs (<10 mN) and limited applications. To reach both the desired forces (>10 N) and displacements (>1 cm), we need to stack multiple layers. Here we build upon our earlier work which combined UV curable strain stiffening elastomers(16) with ultra-thin carbon nanotube (CNT) based

percolative electrodes(17) to make multilayered, robust, completely soft actuators. The use of strain-stiffening elastomers in a multilayered configuration bypasses the need to pre-stretch the elastomer(18), which otherwise negates the advantages of a soft actuator by surrounding it with a rigid frame. Without stiff constraints, DEAs can be made into linear contracting and expanding actuators(19) to mimic the function natural muscle fibers(20) and muscular hydrostats(21), respectively. These linear actuators also serve as simple, yet reliable tools to measure mechanical deformation, and therefore quantify energy density, as well as other physical and electrical properties. The range of strains across different animal muscle types is fairly constant (5-10%), but power and energy densities depend on the type of locomotion(22). For example, the energy density ranges from 0.4 J/kg for a swimming bass at 4 Hz, to 6 J/kg at 25 Hz for the flight muscles in a locust, to 40 J/kg for a running rat at 7.5 Hz(23–25). With the goal of dielectric elastomer artificial muscles that achieve similar energy densities and bandwidths as natural muscles, we aimed for the following combination of actuator capabilities: (i) demonstrate linear contraction or expansion in fully soft DEAs, (ii) match the strain capability and energy density of natural muscle, and (iii) maximize the speed of deformation to reach the viscoelastic limits of the elastomer material.

To achieve these goals, we optimized the electrode composition and multilayer processing to sustain high applied electric fields, without the onset of destructive dielectric breakdown.

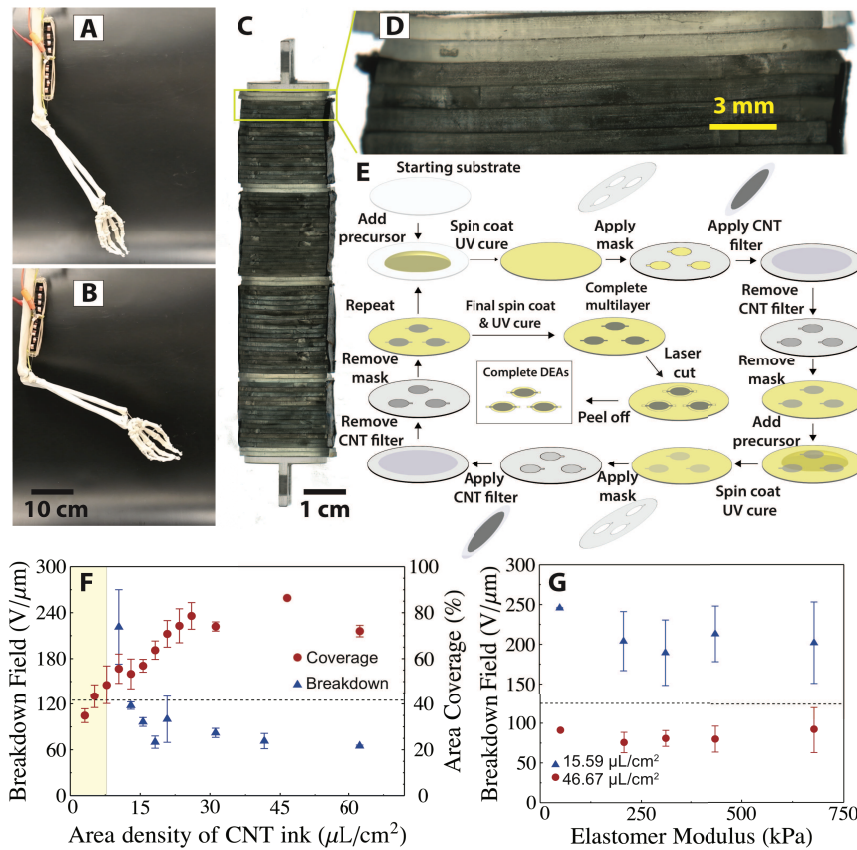
## Significance Statement

To build new robots that can safely interact with people, while completing complex tasks, we need artificial muscles. Ideally, these devices would be completely soft, as strong as natural muscles, and powered by electricity for easy integration with the rest of the robot. We found a materials solution that relies on an established technology, a dielectric elastomer soft capacitor, which deforms when an electric field is applied. By using a unique combination of nano scale conductive particles and soft elastomers, we can apply high electric fields and reach contraction forces on par with natural muscles. Potential uses include novel surgical tools, prosthetics and artificial limbs, haptic devices, and more capable soft robots for exploration.

M.D. designed, fabricated and characterized all types of actuators, with the exception of linear expanding actuators, which were designed by H.Z. E.H. analysed material properties and modeled stress and strain behavior. M.D. collected and interpreted scanning electron microscope images. M.D. analysed data, prepared figures, and wrote the manuscript with comments from all authors. R.J.W., and D.R.C. supervised the project and advised on device optimization.

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**Fig. 1. Fabrication steps and breakdown performance**  
**A,B** Demonstration of muscle-like actuation. A contracting DEA is used to actuate a polymer replica of the bones of a human arm, with mechanics similar to a human bicep muscle. Metal wires serve as rigid links to connect the DEA to the bone. **C**, A contracting actuator showing multiple stacks of active elements (dark grey) alternating with passive elements (light grey) for enhanced adhesion. **D**, Zoomed in image of the top six stacks in the contracting actuator. Each stack consists of twenty five individual dielectric elastomer layers, each approximately 30 microns in thickness, the electrodes are made with ink B at areal density  $15.59 \mu\text{L}/\text{cm}^2$ . **E**, Fabrication sequence for disk actuators used in breakdown testing. **F**, (left y-axis) Breakdown field of the disk actuator as a function of concentration of ink A used. (right y-axis) Estimated areal coverage of carbon nanotubes as a function of concentration of ink used, based on SEM imaging. The results indicate that lower areal densities of CNT reduce the chance of dielectric breakdown. The shaded region left of the  $9 \mu\text{L}/\text{cm}^2$  mark indicates no deformation was observed at any applied field. Dashed line corresponds to the breakdown field when using conventional carbon grease as an electrode determined in house. Each datapoint corresponds to the average breakdown field of five different samples. The error bar corresponds to the standard deviation of the dataset. **G**, Breakdown field as a function of elastomer Young's modulus, at two different areal densities of ink A. The results indicate that the areal density of CNTs dictates the onset of dielectric breakdown, and not the elastomer stiffness. Each datapoint corresponds to the average breakdown field of five different samples. The error bar corresponds to the standard deviation of the dataset.

To understand this motivation in quantitative terms, for small deformations, on par with the strains observed in natural muscles ( $<20\%$ ), the strain in a linear DEA can be approximated as  $s = \epsilon E^2/Y$ , where  $\epsilon$  is the permittivity of the elastomer,  $Y$  is the Young's modulus of the elastomer and  $E$  is the applied electric field. The elastomers used in this work exhibit linear behavior in the range of strains of interest, as demonstrated earlier(17). To first order, the elastic energy density of the deformed elastomer can be approximated as  $U_e = s^2 Y/2$  leading to  $U_e = \epsilon^2 E^4/2Y$ , meaning energy density is highly sensitive to the applied electric field. In addition, we desire thin electrodes, to maximize the volume occupied by the elastomer in the final device.

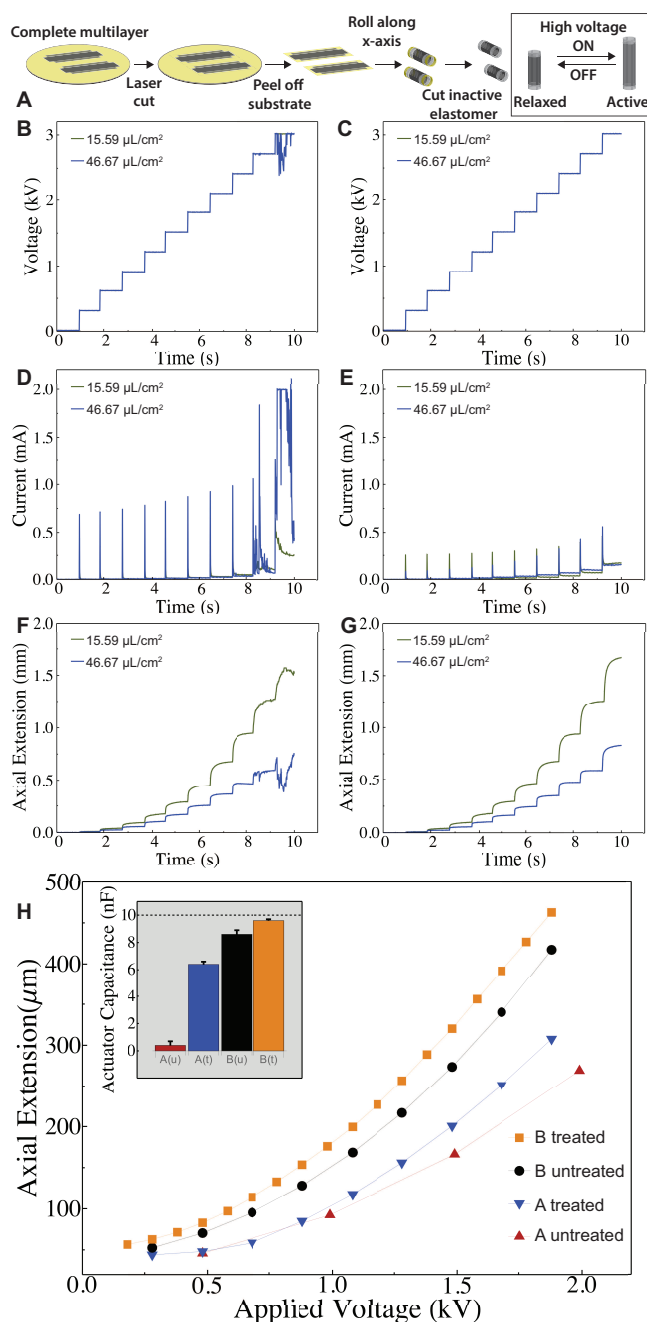
Carbon nanotubes meet the requirement for high conductivity at low thickness(26, 27), and could be easily integrated into a multilayer actuator. Other electrode materials considered were too thick (e.g., hydrogels(28), ionogels(29, 30)), insufficiently conductive (e.g., crumpled graphene(31)), or difficult to integrate in a multilayered actuator (e.g., crumpled metal(32)). The main drawback of carbon nanotubes is the early onset of dielectric breakdown(33) compared to more conventional electrodes, such as carbon grease. Earlier work(34) showed that dielectric breakdown occurs at low strains ( $<20\%$ ), before the onset of strain stiffening in the elastomer. We hypothesized that the reason for early dielectric breakdown is due to non-homogenous distribution of carbon nanotubes causing high electric field concentrations. Based on this hypothesis, we studied the relationship between the areal density of carbon nanotubes and the onset of dielectric breakdown.

Lower areal densities of carbon nanotubes in the electrodes may increase the sheet resistance, which in turn increases the actuator RC time constant. The response speed of DEAs is limited either by how quickly the compliant capacitor is charged, or by how rapidly the elastomer can deform in response to the generated Maxwell stress between the electrodes. We aimed to find electrodes with sufficiently low sheet resistance that the deformation was limited by the elastomer viscoelasticity. As a result, the key actuator performance metrics (i.e., energy density and bandwidth) could be systematically varied by tuning different material properties (i.e., breakdown field and elastomer viscoelasticity, respectively).

## Results

To test the hypothesis that breakdown field depends on nanotube areal density, we fabricated multilayered disk actuators with different CNT amounts, and measured the dielectric breakdown field in each. A schematic of a disk actuator and the multilayering procedure is shown in Fig. 1A. The disk actuators were made by repeated spin coating and UV curing of acrylic elastomer precursors, alternating with stamping of electrodes from a supporting carrier film. The electrodes were made by filtering water-based CNT inks, diluted with isopropanol, through polytetrafluoroethylene (PTFE) filters, used as carrier films. We kept constant the area of the filter ( $9.62 \text{ cm}^2$ ) and ink concentration (determined by optical density, with transmittance equal to  $17\%$  at  $550 \text{ nm}$ ). To systematically modify the CNT areal density, we varied the amount of ink used ( $50\text{--}600 \mu\text{L}$ ), leading to areal densities ranging from  $5.19$  to  $62.37 \mu\text{L}/\text{cm}^2$ . We used two different





**Fig. 2. Performance of roll actuators A.** Fabrication sequence for roll actuators used in CNT electrode optimization and ink selection. **B.** Applied voltage in 0.3 kV steps every 1 s for two roll actuators using ink B, after thermal treatment, at areal densities of 15.59 and 46.67  $\mu\text{L}/\text{cm}^2$ , during the first test of the device. **C.** Corresponding applied voltage during the twentieth test of each device. The noise in the voltage applied to the actuator made with the higher ink concentration is due to high current peaks that reached the 2 mA safety limit of the power supply. **D.** Current response to applied voltage from **B**, during the first test. **E.** Current response to applied voltage from **C**, during the twentieth test. **F.** Actuator axial extension corresponding to **B**. The actuator heights are 1.1 cm for each device. **G.** Actuator axial extension corresponding to **C**. The largest displacement is measured in electrodes made with low areal density of CNT ink, indicating these materials are most suitable for building high performance artificial muscles. **H.** Actuator axial extension as a function of applied field for devices made with four different ink types. The dependence is parabolic: the displacement is proportional to the square of the applied electric field. Inset graph shows the measured actuator capacitance for each ink type. The dashed line corresponds to the calculated capacitance of the device based on geometry and material parameters. The results indicate that ink B after thermal treatment creates the most effective CNT electrode, and should be used in building artificial muscles.

inks: 'A' contained functionalized CNTs dispersed in water (average diameter = 5 nm), while 'B' contained surfactant-coated CNTs dispersed in water (average diameter = 10 nm). The surfactants used for the stable dispersion of purified CNT in ink B at high concentrations in water can be removed at low temperatures by a heating process (2 hours at 120°C), post deposition. The multilayering fabrication process allowed for the electrodes to be heated while on the PTFE carrier films, thus eliminating the need to expose the elastomer to high temperatures. The actuators made with the two inks at varied amounts were tested at increasing electrical fields, as detailed in the Supplemental Materials, until dielectric breakdown occurred.

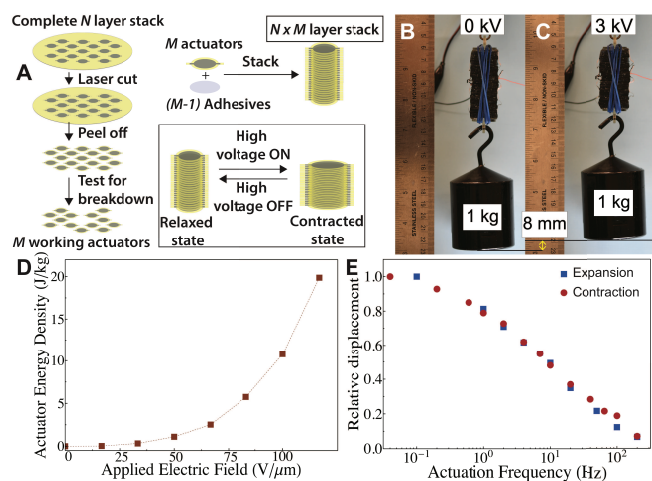
**Carbon nanotube electrode optimization.** The results showed that, for both inks, there is a desirable, low CNT areal density region (7.79-15.59  $\mu\text{L}/\text{cm}^2$ ) in which the breakdown occurs at high electric fields. Reducing the areal density of CNTs increased the dielectric breakdown field to levels higher than more conventional carbon grease electrodes (Fig. 1F). Similar effects were observed for both types of ink used (see SI Appendix, fig. S1A). The onset of dielectric breakdown was found to be approximately inversely dependent with the areal coverage (Fig. 1F). Scanning electron microscopy (SEM) of the CNTs as dispersed on PTFE filters (see SI Appendix, fig. S1 B-J) showed that as the areal coverage of the electrode increases from 30% at an areal density of 5.59  $\mu\text{L}/\text{cm}^2$  to around 70-90% above 20.79  $\mu\text{L}/\text{cm}^2$ . However, the electrodes are not homogenous: patches of dense carbon nanotubes form around 10.39  $\mu\text{L}/\text{cm}^2$ , while some regions remain almost free of CNTs. To our surprise, increasing the elastomer modulus by a factor of 10 $\times$  (through additional crosslinker in the elastomer precursor), did not impact the likelihood of breakdown as much as increasing the areal density of CNTs by a factor of 3 $\times$  (Fig. 1G). SEM imaging of the electrodes on the transfer PTFE films show that the non-homogeneity may be due to aggregation at the filter pores (see SI Appendix, fig. S2A and C). The microstructure is retained after the electrode is transferred onto the elastomer (see SI Appendix, fig S2B and D). Overall, the results indicate that the process gives control over the areal density of the CNTs, which in turn allows the actuators to be powered to higher electric fields than previously demonstrated.

Although a low areal density of CNT electrodes reduces the likelihood of early breakdown, this can lead to poor electromechanical performance due to a low surface coverage of the elastomers. To quantify the effect of CNT areal density on mechanical performance, we built roll actuators<sup>(35)</sup> and measured their linear displacement in the axial direction (Fig. 2A). The expanding actuator consisted of fifteen layers of elastomer (6 cm  $\times$  1 cm), and electrodes with either low (15.59  $\mu\text{L}/\text{cm}^2$ ) and high (46.77  $\mu\text{L}/\text{cm}^2$ ) areal densities of ink B. The actuators were tested by applying increasing voltage steps, while measuring the actuator displacement and current drawn. In the initial test to 2 kV, the higher areal density electrodes drew higher currents than the low areal density electrodes (see SI Appendix, fig. S3 A-C), while showing smaller displacements. In the subsequent test, the same behavior is observed only when the applied voltage is the highest ever applied to the device (2-2.5 kV region, see SI Appendix, fig. S3 D-F). The behavior is repeated when the actuators are charged to 3 kV for the first time (Fig. 2B, D, F). By com-

parison, the current draws are similar at the twentieth cycle (Fig. 2C, E, G), while the displacement is significantly smaller for the high areal density CNT electrodes. The leveling off in the current drawn suggested a self-clearing mechanism was occurring during the initial charge of the actuators(36). Based on our observations and interpretation, the spikes in current cause localized heating and, therefore, partial damage to the electrode network. We expect that the same mechanism is responsible for the complete dielectric breakdown described in Fig. 1. Given that higher current spikes are correlated with higher areal density electrodes, it is likely that the CNT electrode morphology plays a significant role (see SI Appendix, fig. S1). The cause of the current spikes is still unclear: one hypothesis is that the ends of the carbon nanotubes enhance the local electric field. Alternatively, the denser CNT patches may cause excessive heating relative to the more uniform network nearby, or poor bonding between adjacent layers. Given our interest in maximizing actuator performance, we focused on the more robust electrodes made with low areal densities of ink for the remaining demonstrations.

The measured current and applied voltage (Fig. 2) were used to estimate the electrical energy input into the system:  $U_{\text{etotal}} = Q \times V/2 = 200 \text{ mJ}$  for the low areal density actuator, where  $Q$  is the charge stored in the capacitor, as a product of the current applied and time, and  $V$  is the voltage at which the capacitor is charged. The measured displacement ( $d$ ) and blocked force ( $F_b$ ) of the actuator (see SI Appendix fig. S4) were used to estimate the mechanical work output ( $U_{\text{mech}} = d \times F_b/2 = 3.0 \text{ mJ}$  for the low areal density actuator), giving an energy efficiency ( $u_{\text{mech}}/u_{\text{etotal}}$ ) of 1.5%. In addition, we also determined the electromechanical conversion efficiency of the actuator, by comparing the mechanical energy output to the stored electrical energy in the capacitor ( $U_{\text{estored}}$ ). The electrical energy in the capacitor was calculated based on the measured capacitance ( $C = 10 \text{ nF}$ ) and voltage across the capacitor ( $V = 3 \text{ kV}$ ), using  $U_{\text{estored}} = C \times V^2/2 = 45 \text{ mJ}$ . The electromechanical efficiency ( $U_{\text{mech}}/U_{\text{estored}}$ ) of the expanding roll actuator is 6.7%.

To complete the electrode optimization we considered the impact of thermal treatment on the two ink types, for a constant CNT areal density. In addition to measuring the displacement of linear expanding axial actuators, we also quantified the electrode sheet resistance, and the actuator capacitance at low voltage (see description in SI Appendix, fig. S5). We expected that electrodes which are not able to physically cover the entire elastomer surface effectively, would have correspondingly high sheet resistance, lower than calculated capacitance values, and lower actuator displacement. Our expectation was confirmed: the actuators which exhibit the highest displacement also had the highest measured capacitance (Fig. 2F). The best performing material was ink B after thermal treatment (vacuum for 2 h at 120°C), suggesting that the removal of surfactant molecules yields the most effective CNT network. SEM imaging of ink B electrodes showed no significant change in the microstructure before and after thermal treatment (see SI Appendix, fig. S6). Sheet resistance measurements of the inks on PTFE supports are in agreement with this trend (see SI Appendix, table S1). Given the higher breakdown fields ( $> 100 \text{ V}/\mu\text{m}$ ) and large actuator displacements, we selected ink B, after thermal treatment, as the

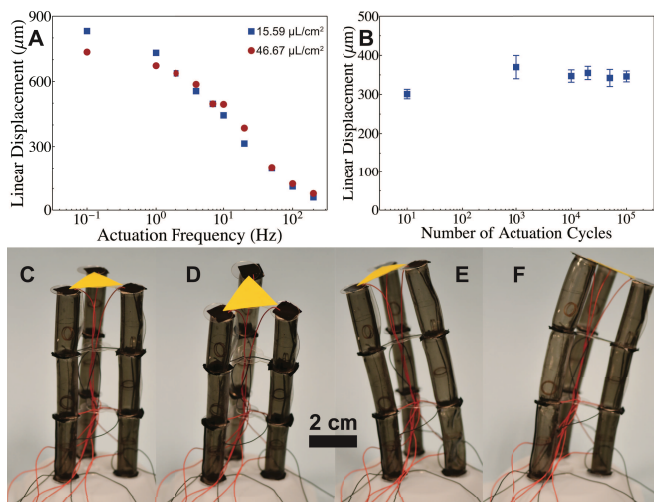


**Fig. 3. Performance of linear contracting actuators** **A**, Fabrication sequence for linear contracting actuators used to measure energy density. **B**, Contracting actuator attached to a 1 kg weight without electrical power applied. The actuator is 6.3 cm long, made of individual  $30 \mu\text{m}$  layers, weighs 20 g, and is made with ink B electrodes, after thermal treatment, at  $15.59 \mu\text{L}/\text{cm}^2$ . **C**, Same actuator powered to 3.5 kV lifting a 1 kg weight by 8 mm. **D**, Energy density of the same actuator from B. The fitted line corresponds to a fourth power dependence of the energy density on the applied electric field, as predicted by our model. At the highest applied field, the actuator exhibits a peak energy density of  $19.8 \text{ J/kg}$ , on par with natural muscle. **E**, Comparison of relative displacements of contracting cylinder and expanding roll actuators as a function of actuation frequency at 3 kV. The contracting actuator was a cylinder: 2 cm long, with a 1.2 cm diameter, made with ink B electrodes, after thermal treatment, at  $15.59 \mu\text{L}/\text{cm}^2$ . The expanding actuator was a cylinder: 2 cm long, with a 1 cm diameter, made with ink B electrodes, after thermal treatment, at  $15.59 \mu\text{L}/\text{cm}^2$ .

optimal electrode for our artificial muscles.

**Artificial muscle demonstrations.** By multilayering our UV curable elastomers and the optimized ink B electrodes, we built a linear contracting DEA to realize our goal of a fully soft, electrically-driven artificial muscle. We adapted an established process(19, 37) by stacking 45 disks each of 26 layers to create a 1170 layer cylinder (see fig. 3A, and SI Appendix Fig. S7 A). To characterize the usable energy density, we measured the weight ( $w$ ), pull force in a mechanical tester with no displacement ( $F_b$ ), and free displacement ( $d$ ) against gravity of the actuator as a function of the applied electric field (see fig. 3B, C, D, and from SI Appendix, fig S7 B, C, D, E, and video S1). The pull force was measured by attaching one end of the actuator to a load cell and holding the other end fixed. The load cell had practically zero displacement, the actuator was powered and the maximum force reading was recorded as the pull force. At high electric fields the energy density ( $= F_b \times d/2w$ ) reached  $19.8 \text{ J/kg}$ , while the displacement corresponded to 24% strain, covering a large segment of the performance envelope of natural muscles(38). For mammalian muscles, the typical energy density is  $8 \text{ J/kg}$ , while the typical strain is 20%. Compared to earlier demonstrations of stacked DEAs, our devices have higher energy density, thinner electrodes (Table 1). Furthermore, our fabrication process allows independent tuning of the properties and processing of the elastomer together with the electrode.

The same building blocks can be used to demonstrate high energy density in linear expanding actuators. A similar performance evaluation of the expanding actuators (low areal den-



**Fig. 4. Limits of performance** **A**, Roll actuator extension as a function of actuation frequency at 3 kV for two different CNT ink compositions. Both expanding actuators were cylindrical: 1.2 cm long, with a 1 cm diameter, made with ink B electrodes, after thermal treatment, at 15.59 and 46.77  $\mu\text{L}/\text{cm}^2$ . The similar performance of different areal densities CNT inks indicate the actuator response is limited by the viscoelasticity of the elastomer and not the RC constant of the circuit. **B**, Roll actuator expansion as a function of number of cycles during continuous testing under a sine wave signal with 10 Hz frequency and 3 kV peak. The expanding actuator was a cylinder: 1.2 cm long, with a 1 cm diameter, made with ink B electrodes, after thermal treatment, at 15.59  $\mu\text{L}/\text{cm}^2$ . The device is robust and can withstand 100,000 cycles without measurable degradation in mechanical performance. Each datapoint corresponds to the average displacement of the same device tested five times. The error bar corresponds to the standard deviation of the dataset. **C, D, E, F** Complex actuators made from simple building blocks: a muscular hydrostat replica made from linear expanding actuators powered to 3 kV. **E** relaxed state, **F** column 1 actuated, **G** column 2 actuated, **H** column 3 actuated.

sities ink actuators from Fig. 2E, using blocked force instead of pull force) demonstrated a peak energy density of 13.75 J/kg, while the displacement corresponded to 22% strain (see SI Appendix, fig. S4). The blocked force was measured by constraining the actuator between a mechanical ground and a load cell that was fixed in place. When an actuator was powered its displacement was fully constrained: the maximum force reading was recorded as the blocked force. The lower energy density exhibited by the roll actuators compared to the contracting cylindrical actuators ones may be due to the mechanical energy lost to expanding the actuators radially. Relative to earlier examples(39), these linear actuators achieve a factor of almost 30 $\times$  higher energy density, primarily due to the higher attainable applied electric field without dielectric breakdown. Despite the different geometry, both the expanding and contracting actuators exhibit similar bandwidth, by comparing displacement relative to the actuator length (Fig. 3E). The results indicate that the device performance is not dictated by the final actuator geometry, but by the properties of the materials used as the building blocks.

## Discussion

To understand the electronic and viscoelastic limits of the device, we characterized the actuator bandwidth (i.e., response time) and electric properties. To this end, we compared two areal densities of ink B after heat treatment (15.59 and 46.77  $\mu\text{L}/\text{cm}^2$ ) on the actuator's linear expansion at a range of frequencies between 0.1 and 200 Hz. The test consisted of ap-

plying a sinusoidal voltage of 3 kV at a given frequency for 1-100 s, and measuring the free displacement. We observed no significant difference in displacement between the two actuators, despite the 3 $\times$  increase in CNT concentration and corresponding 4 $\times$  decrease in sheet resistance. For comparison, the capacitances of the two actuators were similar: 9.6 and 9.9 nF, for the low and high concentrations, respectively. The resistances were 0.6 and 2.2 M $\Omega$ , respectively. Both the bandwidth data and the RC time constants (5-20 ms) indicate the response was limited by material viscoelasticity (Fig. 4A). These actuators did not reach full displacement at frequencies higher than 10 Hz, resulting in a power density on the lower end of the power density in mammalian muscles (100-400 W/kg). Increasing the power density further will require an increase in bandwidth, and materials with lower viscoelastic losses.

For completeness, a 0 to 3 kV 10 Hz frequency signal was applied continuously to explore device robustness. We observed no measurable degradation in performance for actuators made with the low areal density electrodes of ink B (Fig. 4B) over the course of 100,000 cycles of continuous testing. The displacement of the actuators dropped to nearly half the initial value after 300,000 cycles. This fatigue may be due to damage to the carbon nanotube network, which could be reduced by blending with different nanoscale conductors, such as silver nanowires(40). To match the robustness of natural muscles ( $10^9$  cycles), we will need a better understanding of the device degradation. In addition, we used the established simple building blocks (expanding and bending actuators) to create more complex systems with additional capabilities. One example of these complex systems is as a muscular hydrostat replica made from linear expanding actuators, capable of both bending (tilt angles of 30 $^\circ$  in three independent directions) and expansion (10% strain) (see fig. 4C, D, E, F, and SI video S2). We envisioned optimized versions of these types of enhanced actuators could find use in manipulation of delicate objects or other complex manipulation tasks.

The DEAs presented in this work exhibit the highest peak energy density to date, while still being soft and stretchable, without incorporating any liquid components. By carefully selecting and processing ultrathin nanoscale conductors (see SI Appendix, fig. S8) we were able to apply high electric fields without risking premature dielectric breakdown. At high fields, the actuators show strain capability and energy density on par with natural muscles. We found that the actuator speed was limited by the elastomer viscoelasticity, not the electrode sheet resistance. This work represents a major step towards the realization of fully stretchable, electrically driven, strong, and fast artificial muscles to be implemented in a variety of soft machines that can interact with, enhance, or restore the function of the human body.

## Materials and Methods

In brief, we fabricated the artificial muscles using a previously established multilayering technique(17). The artificial muscles were made from UV curable acrylic elastomers and ultra-thin carbon nanotube mats. We quantified the actuator displacement, force output, and bandwidth for both contracting and expanding actuators using laser displacement sensors and load cells. Additional details are provided in the Supplemental Information.



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**Table 1. Performance of dielectric elastomer actuators in comparison to natural muscles.**

| Electrode material       | Natural muscle(22) | CNTs (this work) |           | Carbon powder(37) | Dispersed carbon (19) | Hydrogel(15)        | Corrugated metal(39) |
|--------------------------|--------------------|------------------|-----------|-------------------|-----------------------|---------------------|----------------------|
| Action upon stimulation  | Contraction        | Contraction      | Expansion | Contraction       | Contraction           | Contraction         | Expansion            |
| Pre-strain (x × y)       | N/A                | 1 × 1            | 1 × 1     | 1 × 1             | 1 × 1                 | 3 × 3.5             | 1 × 1                |
| Electrode thickness (μm) | N/A                | <0.1             | <0.1      | 0.5-5             | 3-30                  | 100-200             | 0.1-0.2              |
| Applied Field (V/μm)     | N/A                | 100-150          | 100-150   | 62                | 12                    | 33                  | 35                   |
| Applied Voltage (kV)     | N/A                | 3.75             | 3.75      | 4                 | 6                     | 24                  | 2.8                  |
| Actuation Strain (%)     | 10-30              | 24               | 22        | 30                | 16                    | 69                  | 2.5                  |
| Bandwidth (Hz)           | 2-180              | 1-10             | 1-10      | N/A               | N/A                   | 20                  | 10-100               |
| Max power output (W/kg)  | 9-280              | 30-80            | 30-80     | N/A               | N/A                   | 60-600 <sup>1</sup> | 5-50                 |
| Energy density (J/kg)    | 0.2-40             | 19.8             | 13.75     | 13                | 10                    | 7-70 <sup>1</sup>   | 0.5                  |

<sup>1</sup> Lower bound includes estimated weight of fixtures required to maintain pre-stretch.



## Supplementary Information for

Realizing the potential of dielectric elastomer artificial muscles

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### **This PDF file includes:**

Materials and methods

Figs. S1 to S8

Tables S1

Captions for movies S1 to S2

References for SI reference citations

### **Other supplementary materials for this manuscript include the following:**

Movies S1 to S2

## Materials and Methods

**Material preparation.** The multilayering method was adapted from our earlier work (1). We used the Gent model (2) to select which elastomer formulation to use in our multilayers. The model takes into account the strain hardening behavior of elastomers. The  $I$  value corresponds to the first invariant of deformation tensor at which the strain hardening occurs. The dielectric elastomer must undergo rapid strain stiffening to avoid the electromechanical instability (3). For the Gent model, strain stiffening occurs in materials with  $I$  values lower than seven in the model parameter fit. In order to obtain large actuation strain, we searched through different oligomers and crosslinker concentrations for the elastomer with the lowest modulus that could demonstrate the desired strain stiffening behavior. The elastomer of choice used CN9018 as the oligomer base with 5% hexanediol diacrylate (HDDA) crosslinker, with parametric model fits of  $I=6.97$ , and  $\mu=207.4$  kPa. On one occasion we made elastomers with different crosslinker concentrations (0, 5, 7.5, 10, and 12.5% HDDA) to increase the Young's modulus of the elastomer (50, 200, 300, 450, and 660 kPa, respectively), to evaluate the onset of dielectric breakdown as a function of elastomer stiffness. The electrodes were made using carbon nanotube (CNT) inks A of P3 carboxylic acid-functionalized CNTs from Carbon Solutions Inc. (Riverside, CA) and ink B of surfactant-coated CNTs from NanoC (Westwood, MA). The P3 CNTs were received as powders and suspended in water by ultrasonication (10 minutes at 65% amplitude using an ultrasonic horn, Cole Parmer 500 W Processor, Vernon Hill, IL). All inks were diluted to 17% transmittance at 550 nm (measured in a 1 cm cuvette using a USB2000+ spectrometer from Ocean Optics, Largo, FL) and used as prepared. The diluted inks were mixed with isopropanol then filtered through polytetrafluoroethylene (PTFE) filters (35 or 70 mm in inner diameter, and 47 or 90 mm in outer diameter, respectively, 0.2  $\mu$ m pore diameter from Spectrum, New Brunswick, NJ). The electrode sheet resistance was measured using an Inline Non-contact Sheet Resistance Meter Model 20J3 (Delcom Instruments, Prescott, WI).

**Actuator fabrication.** The acrylic oligomer precursors were spin coated (Laurell Technologies, North Wales, PA) at 6000 RPM for 30 seconds. The resulting thin layers (30  $\mu$ m each) were cured using UV light (Ever Bright Printing Machine Ltd. 365 nm wavelength), under flowing argon gas for 120 seconds. The electrodes were stamped through a laser cut mask made from silicone-coated Mylar for easy release. The spin coating, UV curing, and electrode stamping steps were repeated to reach the target number of active layers. Every device was made with two inactive outer layers to avoid dielectric breakdown from exposed electrodes. The complete multilayer was laser cut to incorporate a 1 mm passive edge and peeled off the PETG wafer. For contraction actuators, multiple disks (between 5 and 50, depending on the experiment) were stacked on top of each other perpendicular to the electrode plane. Passive tension elements (rubber bands) were added on 3D printed connection elements to keep the stack under compression and avoid delamination under large forces. For expanding actuators the multilayer rectangle was rolled along the longer edge to create a roll. Electrical connections were made by cutting the multilayer to expose the electrodes. Colloidal silver (Ted Pella, Redding, CA) was applied to the exposed nanoconductors, then carbon tape (also Ted Pella) covered the silver and connected to wires leading to a high voltage power supply.

**Material and device characterization.** Mechanical properties of the materials are determined following previously established guidelines (4). The dielectric breakdown limit of disk actuators was determined by powering the devices using a Trek 610E power supply and amplifier (Lockport, NY). The current limit was set to 2 mA and voltage was applied as a step function of increasing amplitude, starting at 1 kV and increasing in 0.25 kV increments until breakdown was

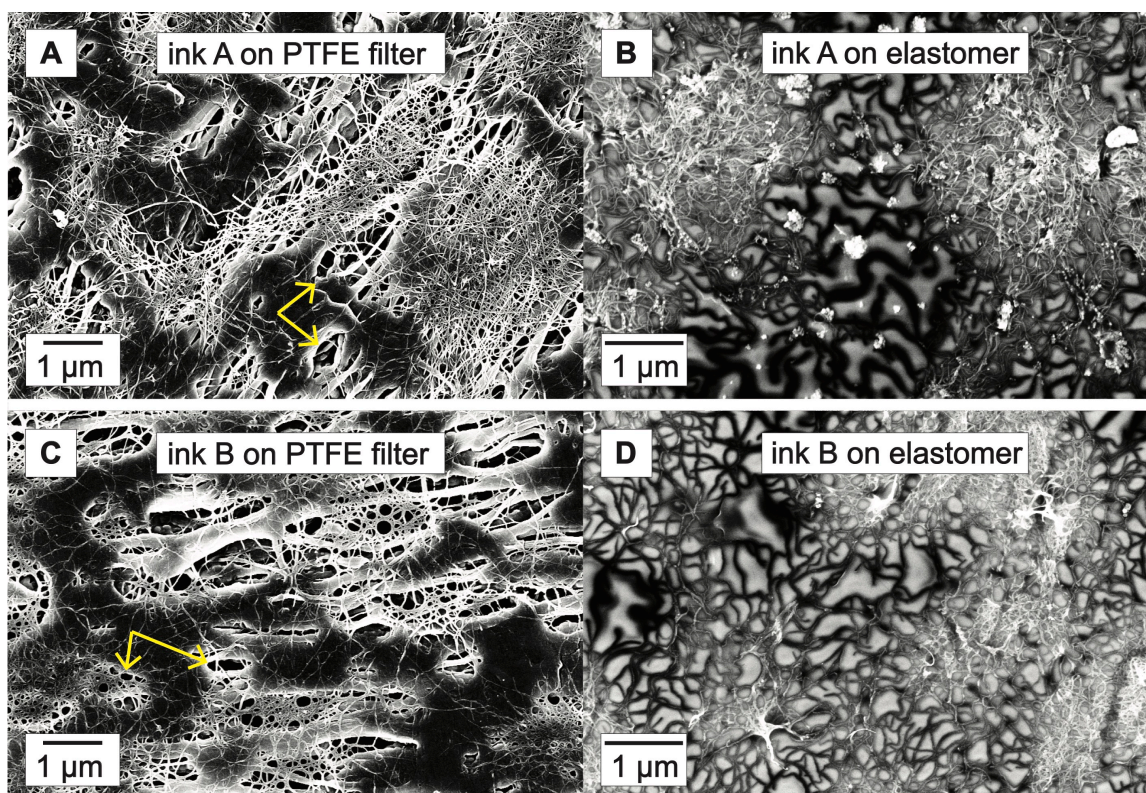


observed. Additionally, the linear actuators were used to determine the current and displacement responses during early testing at high voltage. The linear expanding actuators were secured at the bottom of the roll and powered using the high voltage power supply. The current at the power supply was recorded and the upward displacement was measured using an MTI 2100 Fotonic Sensor (MTI Instruments, Albany, NY). Similarly, the blocked force was measured using a Nano 17 load cell (ATI, Apex, NC) mounted above the actuator. Linear contracting actuators were characterized in an Instron force tester (Norwood, MA). The pull force was measured for a clamped actuator, powered to high voltage. The bending hydrostat actuators were powered using the high voltage power supply and imaged using a Nikon camera. The photographs were analyzed using image processing software (imageJ) to determine tip displacement.

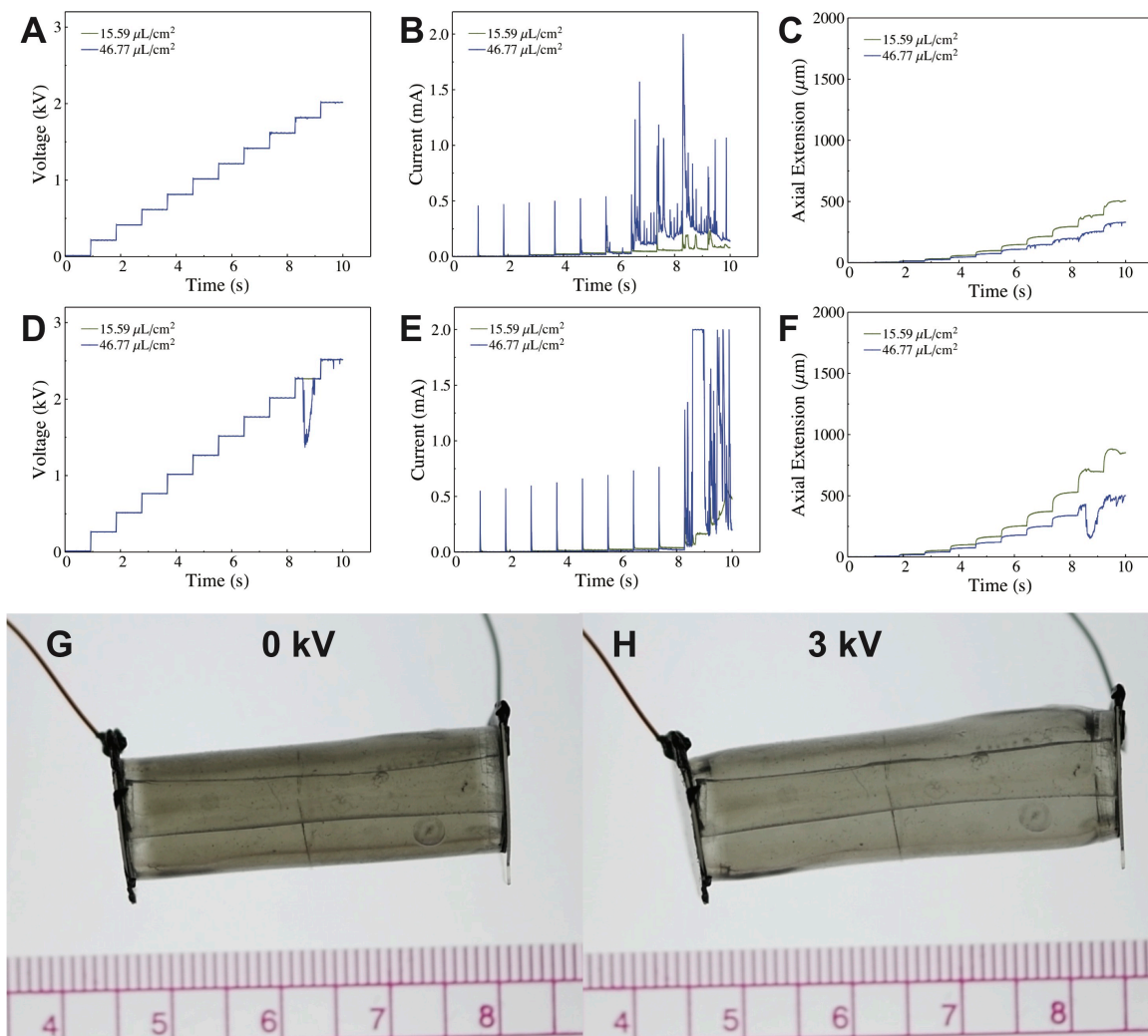
**Material and device imaging.** Electrodes were imaged using an Ultra 55 Scanning Electron Microscope (Zeiss, Oberkochen, Germany). The electrodes were transferred from the PTFE filters onto carbon tape (Electron Microscopy Sciences, Hatfield, PA). The micrographs were analyzed using imaging processing software (imageJ) to obtain numerical data for area coverage. Photographs and supporting videos are acquired with a digital DSLR camera (Nikon D7500, Nikon Corp, Japan). Video sequences are clipped from raw footage and exported using iMovie (Apple Corp, Cupertino, CA, USA).





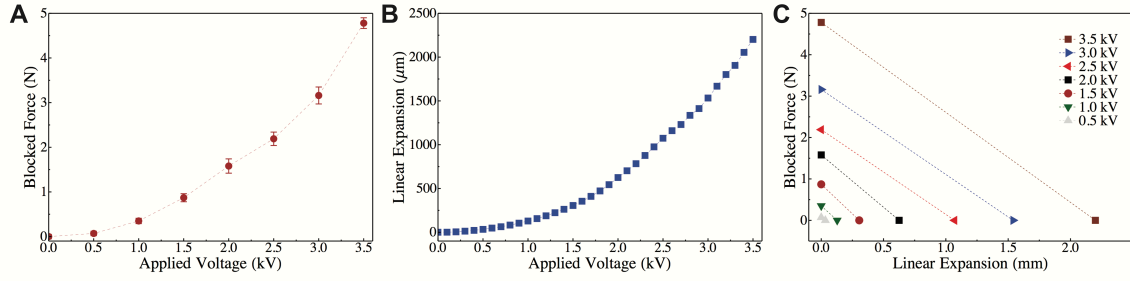


**Fig. S2. Electrodes on PTFE and elastomer substrates.** **A**, SEM image of carbon nanotubes at  $15.59 \mu\text{L}/\text{cm}^2$  (ink A, post thermal treatment) on the PTFE transfer filter. The image shows aggregation of the carbon nanotubes is more prevalent near the filter's pores. Yellow arrows are used to indicate the edges of pores to help distinguish the membrane microstructure from the agglomerations of carbon nanotubes. **B**, SEM image of carbon nanotubes at  $15.59 \mu\text{L}/\text{cm}^2$  (ink B, post thermal treatment) on the PTFE transfer filter. The image shows aggregation of the carbon nanotubes is more prevalent near the filter's pores. **C**, SEM image of carbon nanotubes at  $15.59 \mu\text{L}/\text{cm}^2$  (ink A, post thermal treatment) on the acrylic elastomer. Yellow arrows are used to indicate the edges of pores to help distinguish the membrane microstructure from the agglomerations of carbon nanotubes. **D**, SEM image of carbon nanotubes at  $15.59 \mu\text{L}/\text{cm}^2$  (ink B, post thermal treatment) on the acrylic elastomer. The image is distorted due to the non-conductive substrate, but overall shows a similar pattern of disperse and concentrated patches of carbon nanotubes. The electrodes on PTFE were sputter coated with copper (10 second deposition, 600 mA, 400 V) using a Denton Vacuum Pro Desktop Coater (Moorestown, NJ).

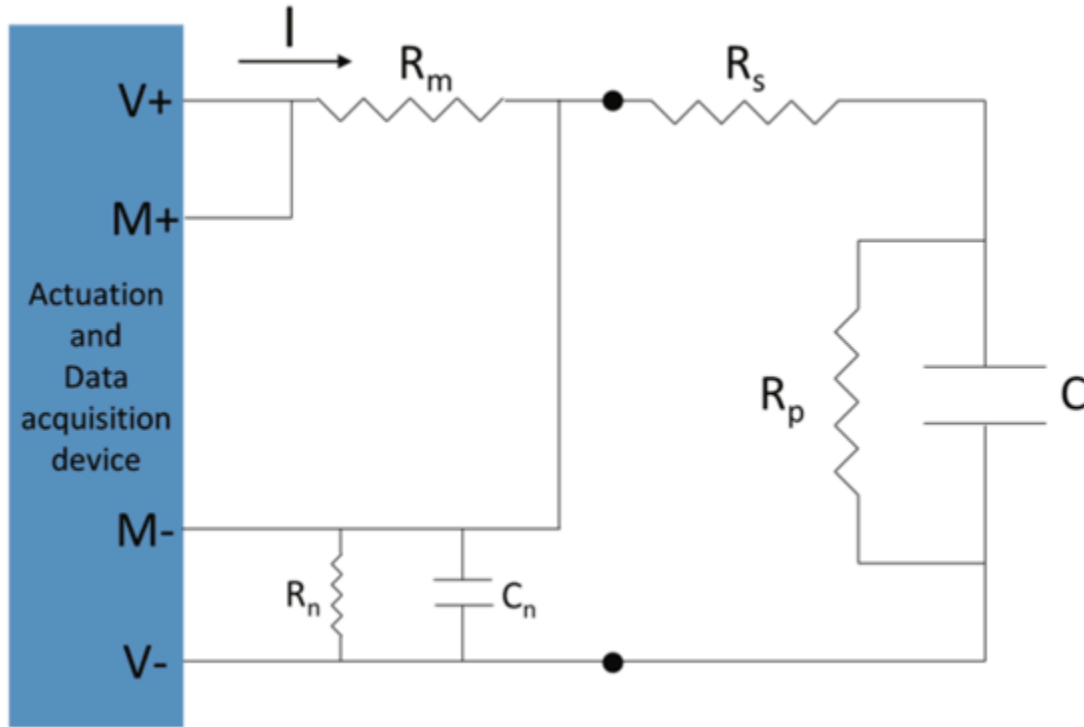


**Fig. S3. Ink selection experiments.** A. Voltage applied to two axial extending roll actuators up to 2 kV. Actuators using two different ink B carbon nanotube area densities (15.59  $\mu\text{L}/\text{cm}^2$  and 46.77  $\mu\text{L}/\text{cm}^2$ ) B. Current response to applied voltage in A. C. Displacement of the actuators recorded in response to applied voltage in A. D. Voltage applied to two axial extending roll actuators up to 2.5 kV. Actuators using two different ink B carbon nanotube area densities (15.59  $\mu\text{L}/\text{cm}^2$  and 46.77  $\mu\text{L}/\text{cm}^2$ ) E. Current response to applied voltage in D. F. Displacement of the actuators recorded in response to applied voltage in D. G, Example of an axial extending roll actuator in its relaxed state. The actuator was made with 15.59  $\mu\text{L}/\text{cm}^2$  ink B electrodes, after thermal treatment. H, The same roll actuator powered to 3 kV.

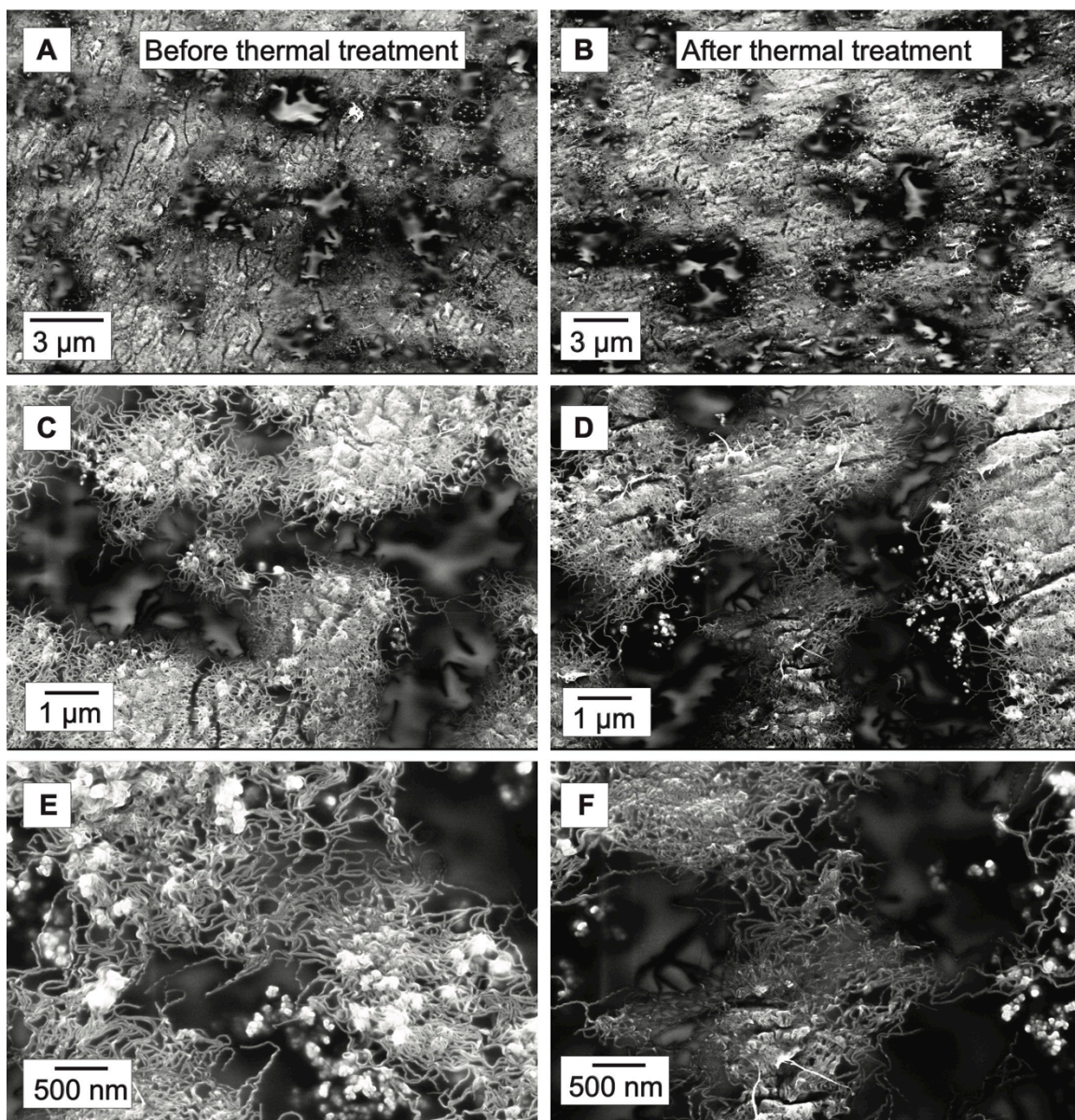




**Fig. S4. Roll actuators.** **A**, Measured blocked force as a function of applied voltage for a 0.4 g actuator with 30  $\mu\text{m}$  layers, with ink B electrodes, after thermal treatment, with an area density of  $15.59 \mu\text{L}/\text{cm}^2$ . **B**, Measured axial extension as a function of applied voltage for the same 0.4 g actuator with 30  $\mu\text{m}$  layers. **C**, Blocked force and axial extension measurements at different voltage steps for the same 1.2 cm tall, 0.4 g actuator with 30  $\mu\text{m}$  layers. The dashed lines guide the eye to show the mechanical energy output of the actuator as the area under the curve. At the highest applied voltage, an energy density of 13.75 J/kg, on par with natural muscle, is achieved.

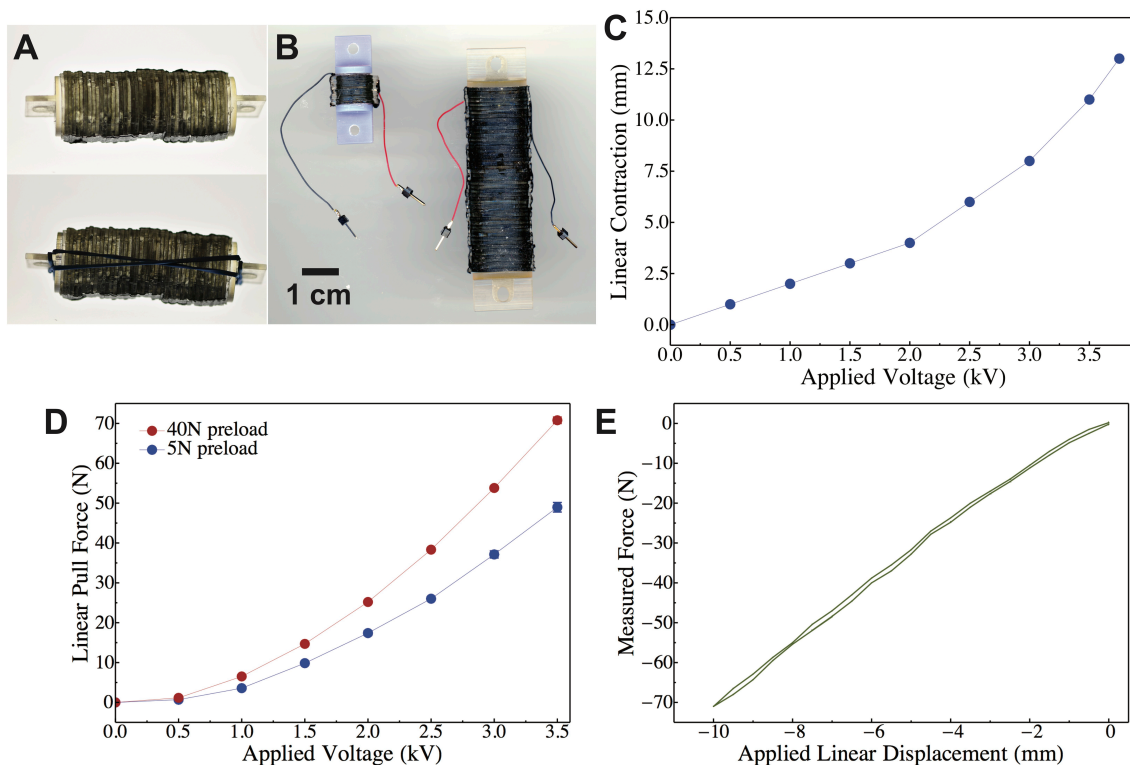


**Fig. S5. Capacitance measurement setup.** Schematic of the circuit used to measure low voltage (10 V) capacitance of the linear expanding actuators for CNT ink selection. To measure the current input into a capacitance, we employ a known series resistance of 1 MOhm. The voltage across this measurement resistance ( $R_m$ ) is monitored to obtain series current going to the capacitance. An algorithm is implemented to switch on  $V_{in}$  and monitor the instantaneous current through  $R_m$ , such that the leakage current and accumulated charges can be obtained. The capacitance value is measured for the no-load case, while keeping the leads of the measurement circuit disconnected. The measured capacitance for this case is noted as  $C_n$  and the leakage resistance value is noted as  $R_n$ . These measured values are the resistance and capacitance associated with the connector leads and internal circuit of the measurement setup. Once these values are obtained, the capacitance system in question is connected and the experiment is repeated. The noise values are subtracted from the experimental measurements and the capacitance values are corrected for series and parallel resistance.

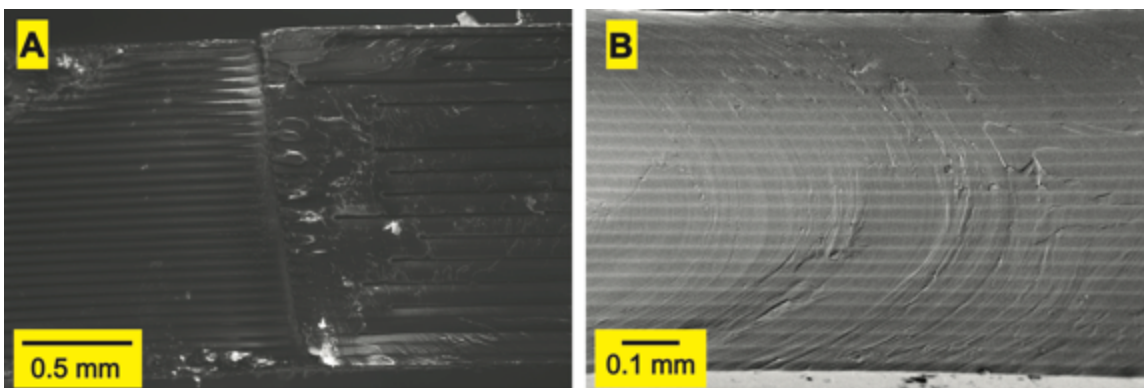


**Fig. S6. Impact of thermal treatment** A, C, E, SEM images of ink B CNTs transferred onto carbon tape directly from PTFE filter, at  $12.99 \mu\text{L}/\text{cm}^2$  before thermal treatment. The three images correspond to magnifications of 10k, 30k and 60k X. B, D, F, SEM images of ink B CNTs transferred onto carbon tape directly from PTFE filter, at  $12.99 \mu\text{L}/\text{cm}^2$  after thermal treatment ( $120^\circ \text{C}$  for 2 hours under vacuum). The three images correspond to magnifications of 10k, 30k and 60k X. The similarity between the untreated and treated samples suggests the increase in conductivity is not due to changes in morphology.





**Fig. S7. Linear contracting actuators.** **A**, Example of a linear contracting actuator without (top) and with (bottom) a passive tension (rubber band) element which keeps the individual components in close contact. **B**, Examples of different size actuators: left is a 1 g device in which the passive elements make up 50 % of the entire actuator weight; right is a 20 g device in which the passive elements make up <10 % of the entire actuator weight. **C**, Measured free linear contraction as a function of applied voltage for a 20 g actuator with 30  $\mu\text{m}$  layers. The electrodes were made of ink B, after thermal treatment, with an area density of  $15.59 \mu\text{L}/\text{cm}^2$ . The initial length of the actuator was 6.3 cm. **D**, Measured pull force as a function of applied voltage for the same 20 g actuator with 30  $\mu\text{m}$  layers starting with either 5 or 40 N of compression. **E**, Measured force required to deform the same 20 g actuator with 30  $\mu\text{m}$  layers in a mechanical testing apparatus (Instron).



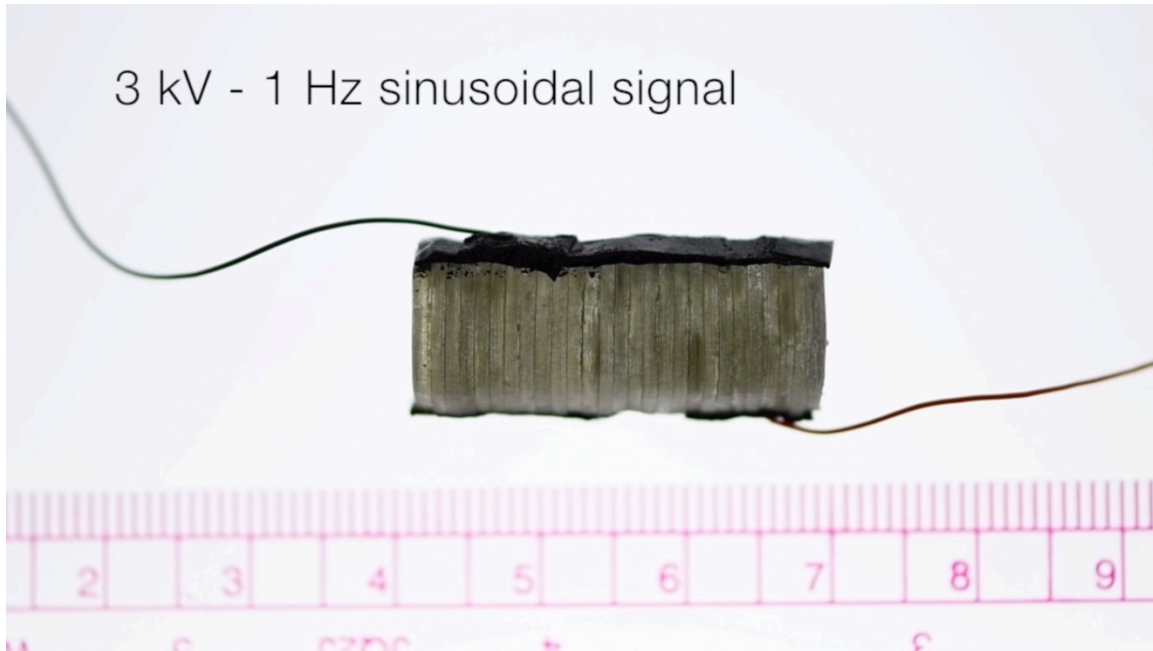
**Fig S8. Cross section imaging.** **A**, SEM image of a disk actuator used in the fabrication of a linear contracting device. The region on the left corresponds to the laser cut edge of the actuator and shows each layer has a thickness of 30  $\mu\text{m}$ . The region on the right corresponds to the edge cut with a blade to make electrical connections. The darker regions every other layer correspond to areas in which CNT electrodes are placed. The electrodes were made of ink B, after thermal treatment, with an area density of 15.59  $\mu\text{L}/\text{cm}^2$ . **B**, SEM image of a bending actuator used in the fabrication of a linear contracting device. The entire region corresponds to the laser cut edge of the actuator and shows each layer has a thickness of 13  $\mu\text{m}$ . The electrodes were made of ink B, after thermal treatment, with an area density of 46.77  $\mu\text{L}/\text{cm}^2$ .

**Table S1. Sheet resistance of different inks measured on PTFE support (kΩ/sq).**

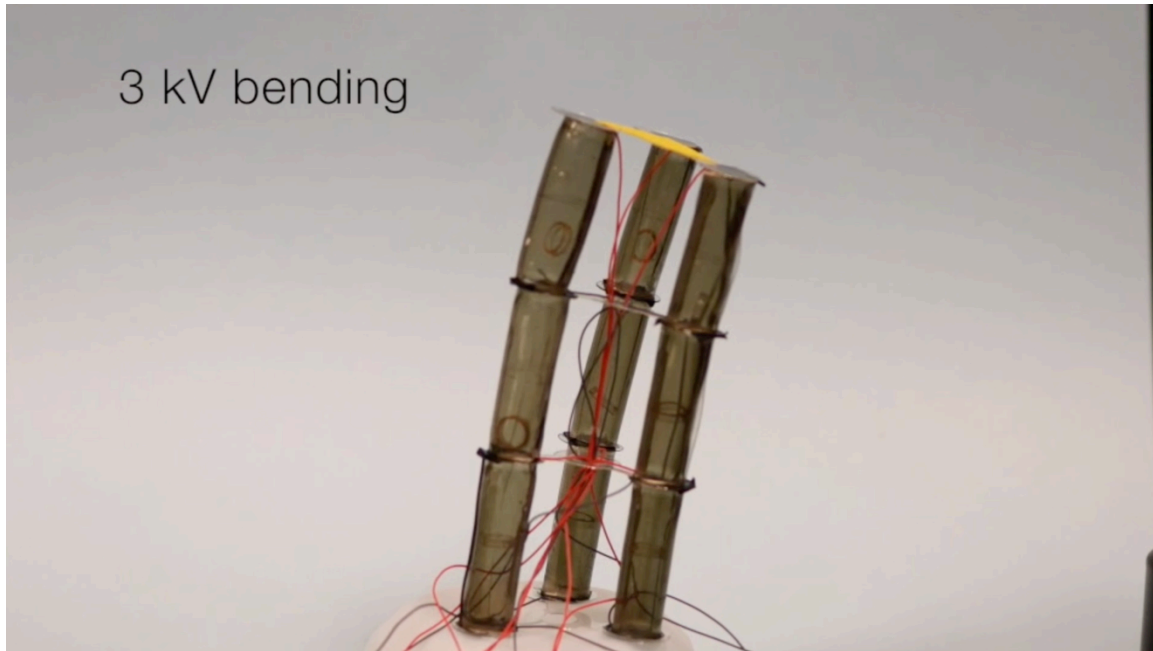
| Area density of CNTs (μL/cm <sup>2</sup> ) | A untreated | A treated | B untreated | B treated |
|--------------------------------------------|-------------|-----------|-------------|-----------|
| 10.4                                       | >100        | >100      | >100        | >100      |
| 20.8                                       | >100        | 100       | 90          | 40        |
| 41.6                                       | 30.0        | 20.0      | 10.0        | 9.4       |
| 62.4                                       | 7.3         | 6.7       | 4.4         | 4.0       |
| 103.9                                      | 2.5         | 2.2       | 1.7         | 1.5       |



**Movie S1.** Schematic description of multilayering process. Schematic description of stacking multilayer disks to assemble a linear contracting actuator. Demonstration of actuation of a linear contracting actuator on a flat light bed at 3 kV and increasing frequency (1, 4, 7, 10 Hz). Demonstration of a linear contracting actuator lifting a 1 kg mass. Demonstration of an artificial muscle powered at 3 kV lifting a human skeleton replica forearm. The mass of the actuator is 80 grams, corresponding to approximately the quarter of a bicep found in a natural system.



**Movie S2.** Schematic description of fabrication of an expanding roll actuator. Demonstration of actuation of an expanding roll actuator on a flat light bed at 3 kV and increasing frequency (1, 4, 7, 10 Hz). Demonstration of an artificial muscular hydrostat made of three columns of linear expanding actuators. The first mode of actuation is bending by powering a single column of actuators at 3 kV. The second mode of actuation is linear expansion by powering all columns simultaneously at 3 kV.



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