

Multidirectional Artificial Muscles from Nylon

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There has been a great advancement in the field of muscle-like actuators (i.e., artificial muscles) since the discovery of new emerging materials such as thermoresponsive polymers,^[1] conducting polymers,^[2–4] dielectric elastomers,^[4–6] carbon nanotubes,^[7–10] graphene,^[11–13] and other materials. Enhancing the performance metrics such as cycle life, gravimetric/volumetric energy and/or power density, efficiency, cost, and controllability of artificial muscles has been an active field of research. Yet there are few artificial muscles that have a good combination of the mentioned performance parameters. For example, although shape memory alloys can provide high contractile stress (200–480 MPa), their poor cycle life (<1000 cycles) at high tensile strains, high cost (300 \$ kg⁻¹), low efficiency, and difficult controllability limit their applications. Conducting polymers offer contractile stresses of 2–120 MPa and require low excitation voltage (<4 V), but they are slow, suffer from low electrochemical coupling, and also scalability of their produced force in the current generation of small film actuators is on the order of few mN.^[4,14] The recently discovered miniature linear and torsional artificial muscles fabricated from carbon nanotube yarns^[8,9] and nanowire yarns^[15] offer long cycle life (>1 million cycles), but their manufacturing cost is not low enough for commercialization yet. Recently nylon fibers (e.g., like those used as high performance fishing lines and sewing threads) have been introduced to the field of artificial muscles.^[1,16] Nylon, when highly oriented along its length, has anisotropic thermal expansion behavior which enables relatively large linear tensile actuation (up to 49%) when the fibers are highly twisted and coiled.^[1,16] These inexpensive (5 \$ kg⁻¹) high-strength polymer fibers can lift loads over 100 times heavier than that of human muscle with the same length and weight.^[1] This combination of 49% tensile actuation and 2.63 kJ kg⁻¹ energy density^[1] opens the door for many biomimetic applications. For example, artificial fingers inspired from human hand anatomy are made from twisted-coil nylon filaments.^[17,18] One of the drawbacks of these bending artificial muscles is that they are converting the linear motion of the twisted-coil nylon fibers to a bending motion which requires a large space for storing the linear actuator. In this work, we have taken a completely different approach to make bending and multidirectional artificial muscles with nylon. Our approach uses the nylon actuator itself to bend and thus eliminates the need for a mechanical transmission mechanism and space for storing long linear actuators which makes their scalability and controllability much easier.

Here, we demonstrate that by roller-pressing highly oriented nylon filaments into rectangular or square cross-section beams and applying heat to one side of those structures, a bending or multidirectional artificial muscle can be made (Supporting Information).

With rectangular-cross section nylon beams fully reversible bending in one plane (X or Y) was achieved, while nylon square rods enabled 2D bending (XY planes) when heat was applied to different sides of the body. The required heat for actuation was generated via two mechanisms: Joule heating and photo-thermal heating. To create the Joule heating traces on the surfaces of the nylon beams, we developed a conductive paint which is a colloidal suspension of micrometer-size silver flakes mixed with metallic nanowires, such as niobium,^[19] in an adhesive resin (Figure 1A–F) (Figure S2, Supporting Information). Any type of highly conductive nanowire with appropriate length (e.g., few mm) should work as well. For the photo-thermal excitation, to increase the light absorption at the surfaces of the actuator, we dip-coated a nylon beam with a thermally conductive paint solution that we developed from high thermally-conductive graphene flakes (Figure 1C) and then actuated the beam with two high power laser diodes (Figure S3, Supporting Information).

Oriented nylon fiber has an anisotropic thermal expansion behavior; it shrinks in length by about 2.5% and expands in thickness by about 4.5% from 25 to 140 °C (Figure 1I). This negative thermal expansion coefficient originates from the fact that amorphous phases of highly drawn polymers such as nylon are energetically less stable than their crystalline phases. Therefore, they can provide large reversible contraction upon accessing conformational entropy when heated (Figure S11, Supporting Information).^[1,20] As Figure 1I suggests, the lateral thermal expansion coefficient changes nonlinearly as a function of temperature from $<-2.8 \times 10^{-5} \text{ K}^{-1}$ to a large negative thermal expansion coefficient of $>-2.8 \times 10^{-4} \text{ K}^{-1}$ above the glass transition temperature (T_g). This negative thermal expansion behavior is the governing phenomenon behind the nylon actuators' working principle. In thermally activated bending actuators, such as thermal bimorphs, there usually is a thermally insulating layer between the top layer and bottom layer to maintain the thermal gradient needed for actuation. Because of the poor thermal conductivity of nylon, we used only one rigid body in the design of our actuator without any thermal insulator layer. As Figure 1H suggests, thermal conductivity of the nylon precursor that we used is around $0.1 \text{ W m}^{-1} \text{ K}^{-1}$ which is very low compared to silver ($\approx 420 \text{ W m}^{-1} \text{ K}^{-1}$) for example. Therefore, according to the heat diffusion time constant equation (Equation (1)), it takes up to several seconds for the heat to transfer across the thickness of the nylon beam. This slow heat transfer rate provides enough time for the heated part to contract and bend the entire beam (Figure 2B).

$$\tau \sim \frac{t^2}{4\alpha} = \frac{\rho t^2 C_p}{4k} \quad (1)$$

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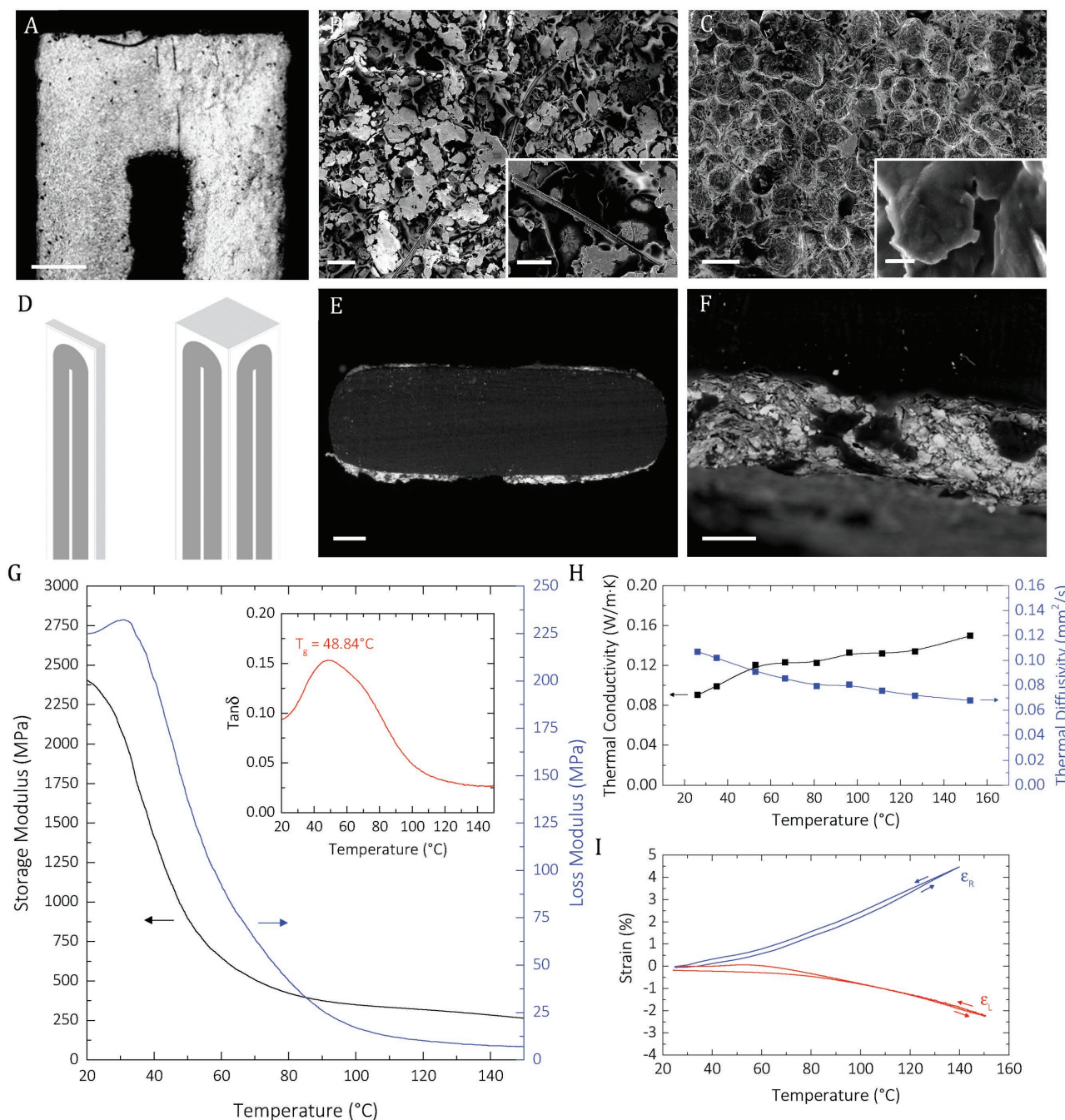


Figure 1. (A) A scanning electron microscope (SEM) image of the patterned conductive coating on a nylon beam (scale bar 400 μm). (B) The SEM image of the conductive paint shows the silver flakes and the nanowires (scale bar 5 μm). The inset shows one individual Nb nanowire mixed with silver flakes and the resin (scale bar 2 μm). (C) A SEM image of the graphene-based thermally conductive coating developed for the photo-thermal excitation of the actuator (scale bar 30 μm). The inset is a small clump of graphene agglomerated (scale bar 300 nm). (D) Schematic of the bending (left) and multidirectional (right) actuators made from nylon beams and square rods. The gray paths represent the Joule heating traces made from the conductive paint. (E) and (F) SEM images of the cross-section of the bending actuator show coating thickness of 40 μm . Scale bars 250 μm and 25 μm respectively. (G) Dynamic modulus as a function of temperature for an annealed nylon beam (l,w,t: 9.69 mm, 1.88 mm, 0.249 mm). The inset shows the $\text{Tan}\delta$ (with δ representing the phase lag between stress and strain). Maximum of $\text{Tan}\delta$ occurs at 48.8 °C which also can be interpreted as the glass transition temperature. (H) The thermal conductivity and diffusivity of nylon as a function of temperature. (I) Thermal expansion across the thickness (ϵ_R) and length (ϵ_L) of a nylon beam as a function of temperature.

where t is the thickness, α is the thermal diffusivity, k is the thermal conductivity, ρ is the volumetric mass density, and C_p is the specific heat capacity.

Another phenomenon that helps the bending is the sensitivity of nylon's dynamic modulus to temperature change. By increasing the temperature from 25 to 150 °C, the storage

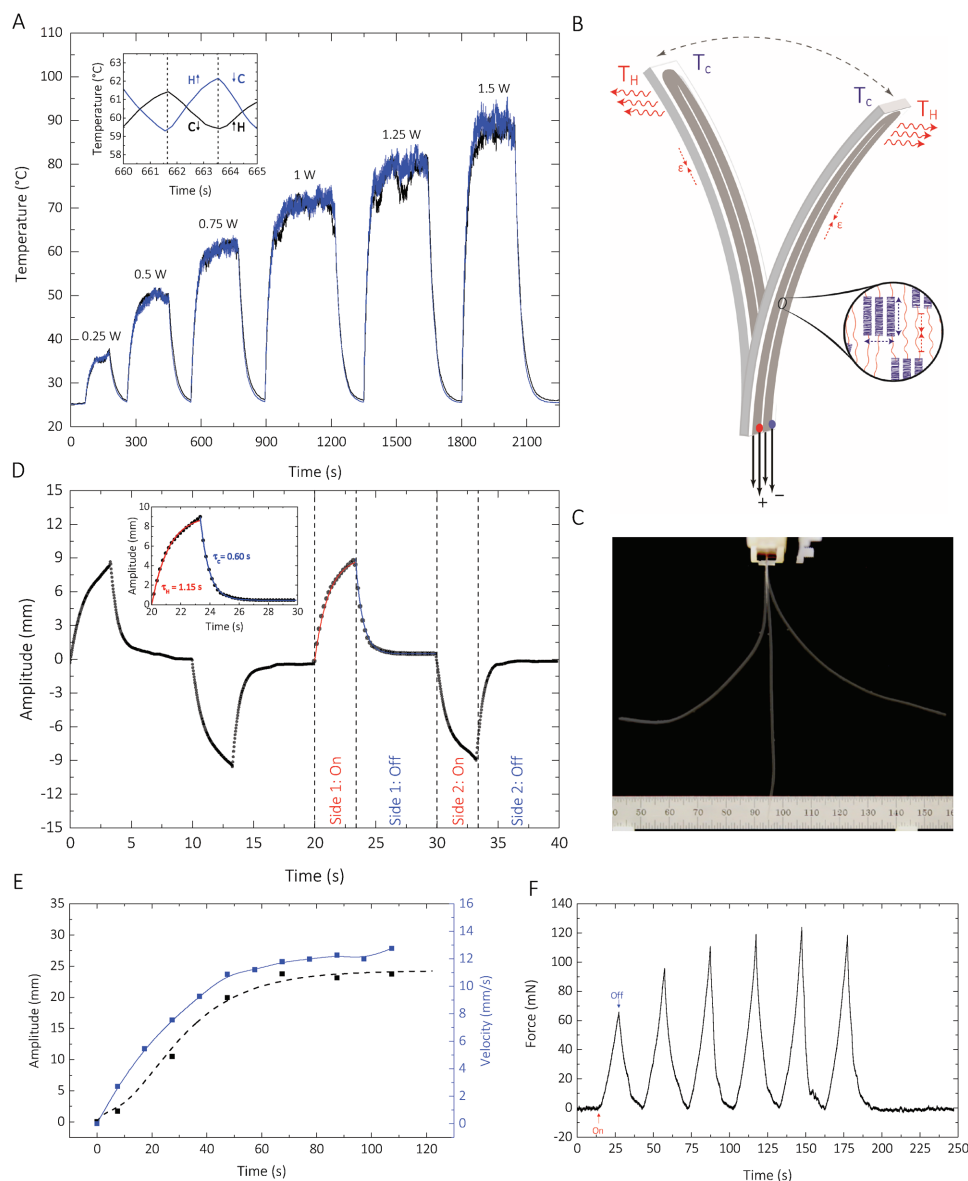


Figure 2. A) Temperature as a function of time for a bending actuator excited at constant power continuous square wave input signal. At each input power the temperature reaches a plateau after several excitations. The inset shows a temperature gradient of 3 °C across the thickness of the actuator at input power of 0.75 W. B) By heating one side of the actuator, the amorphous chains (red lines) shrink in length and the crystalline regions (blue lines) expand in volume. The result is surface contraction of the beam at its heated surface which creates the bending motion. C) Image of a nylon beam reversibly bending. D) Discrete excitation of an actuator (l,w,t: 66.6 mm, 3.54 mm, 0.64 mm) in the format of “on-off-wait” at constant power. The data from 20 to 30 s is under-sampled by a factor of 6 to illustrate the fit better. Inset: the cooling and heating of the actuator can be modeled by an exponential response. For bending we have $A = A_0[1 - \exp(-t/\tau_H)]$ and for relaxation we have $A = A_0 \exp(-t/\tau_C)$, where A_0 is the amplitude of actuation from the zero position, τ_H and τ_C are the time constants for heating and cooling, respectively. The τ_H and τ_C are 1.15 s and 0.60 s, respectively. E) Amplitude and the peak velocity of a bending actuator with continuous excitation as a function of time. The model fits with the experimental results (dashed line vs square points). F) Generated force by a nylon bending actuator (l,w,t: 80 mm, 2.6 mm, 0.7 mm) which was annealed at 150 °C in response to an input power of 2 W (9.6 kW m⁻²).

modulus (ϵ') of nylon drops by 88% (Figure 1G) which makes it easier for the contracting side to bend the whole structure. This large drop in the modulus is also responsible for the decay in the blocking force generated at the tip of the actuator at high temperatures ($F_{bl} \sim \epsilon'$). The $\tan \delta$ plot (Figure 1G inset) suggests a glass transition temperature of around 49 °C for the nylon precursor that was used in this work, which is in agreement

with the value that was measured from differential scanning calorimetry technique (Figure S5, Supporting Information).

A fully recoverable symmetric dynamic range of 1.25 (peak-to-peak amplitude normalized to the length of the actuator) at input power of 19.4 kW m⁻² (or mW mm⁻²) was measured, which is close to the theoretical limit of 1.2732 (i.e., $4/\pi$) for bending actuators (Figure 2C) (Appendix and Video S1,

Supporting Information). Reversible radius of curvature of 53.6 mm was achieved from this sample which agrees to what the model suggests at temperatures of around 96 °C. This radius of curvature is smaller than the 83.3 mm reported for spongy graphene-based bimorph actuators,^[12] but larger than the 10 mm reported for trilayer polymer actuators.^[4] Radius of curvature of 21.6 mm was achieved at input power of 21.55 kW m⁻² but the nylon beam slightly deformed permanently in shape. For a temperature range of 25–150 °C our model suggests a lower bound of 16.2 mm for the radius of curvature (Supporting Information).

By alternating the input power between the two sides of the actuator, we observed that the surface temperature of the actuator increases until it reaches a plateau which itself is a function of power (Figure 2A) (Figure S8, Supporting Information). In addition to the temperature we observed that the amplitude also grows over several cycles until it reaches a plateau (Figure 2E). As Figure 2A (inset) suggests, at the plateau, a temperature gradient of around 3 °C keeps the beam actuating. This temperature gradient is small compared to the thermal gradient required for phase transition materials such as VO₂ ($\Delta T = 15$ °C).^[21] It is important to note that at very long pulses the amplitude can reach the plateau within even one cycle.

The behavior of the nylon was further investigated by applying power in the form of “pulse side one – wait – pulse side two – wait” instead of continuously actuating the muscle in form of “pulse side one – pulse side two”. The waiting periods were long enough to ensure thermal equilibration of the actuator at room temperature. As Figure 2D shows we did not achieve a growing amplitude. The advantage here is that the steady-state response is not spread over several cycles and within one cycle we can achieve the desired amplitude.

Aside from the outstanding displacement performance, nylon bending actuators can also generate a remarkable amount of blocking force even at only 12% of the storage modulus value at room temperature. We measured normalized forces (Supporting Information) of up to 7.5 MPa (fully recoverable—no catch) (Figure 2F) and 55 MPa (with catch-index of 0.12) (Figure 4B) from the bending actuators (Figure S7, Supporting Information). These measured forces are higher than the normalized peak force reported for an ionic-liquid-based bucky gel (231 kPa),^[22] conducting polymer bending actuators (0.4–0.9 MPa),^[14,23] and IPMC-based bending artificial muscles (2–3 MPa).^[24] The measured fully recoverable force is comparable to the normalized blocking force of 6.8 MPa generated by short piezo bimorphs (i.e., <10 mm), but the full dynamic range of these piezo bimorphs is only 400 μ m (0.04 normalized to the length) at zero tip load.^[25]

The measured peak force is lower than the peak force that can be achieved from a nylon linear actuator (in form of a straight filament or twisted-coil fiber). This difference can be explained by the mechanics of materials—bending artificial muscles produce large displacements per unit length of the muscle but the force they can generate is smaller than what they can generate in the form of a linear actuator.

As mentioned previously, it is very critical to maintain a temperature gradient across the thickness of the beam. Beam thickness and the ambient temperature (in a passive cooling setup) are two of the parameters that can affect the bending amplitude

(Equation (2)). By decreasing the temperature gradient, the bending actuator turns into a linear actuator which can create a large amount of force and very small displacement orthogonal to the axis of the actuator.

To compensate for any variations in resistance of the Joule heating traces during the actuation or the fabrication process, we controlled the applied voltage by a PID controller to ensure a constant power is delivered to both sides of the actuator during actuation (Figure 3A) (Supporting Information).

By cycling the input power, we observed a fully reversible amplitude response (Figure 3B,C). This reversibility is very helpful in controlling the position of the actuator's tip without using any position sensors.

To explain the behavior of the actuator, we developed a mathematical model that relates the length of the actuator and other parameters to the displacement at the tip of the actuator as follows (Supporting Information):

$$\delta = -\alpha_T \frac{\Delta T}{2h_T} L^2 \quad (2)$$

where α_T is the temperature-dependent coefficient of thermal expansion, ΔT is the difference between the temperature at the surface of the actuator (T) and the initial temperature (T_i) (e.g., temperature gradient across the thickness), h_T is the thickness of the actuator which is temperature-dependent, and L is the length of the actuator. Since α_T is temperature-dependent, the displacement at the tip of the actuator is nonlinear with respect to the temperature change. Figure 3C,D confirms this nonlinear relationship. By using the values measured for the thermal expansion coefficients (Figure 1I) and the slope of the temperature and power relationship (Figure 3D), we can predict the value of the amplitude as a function of temperature (Figure 3D and Figure S10A, Supporting Information). As shown in Figure 3D, the modeling results (black dashed line) follow the trend in the experimental data (black squares).

During the annealing process, nylon shrinks in length by up to 25%. We observed that as we decrease the annealing temperature from 150 to 60 °C, the reversibility of the actuation motion will also decrease. In other words, the bending actuator can remain in a fixed location after turning the actuator off. This is analogous to the ‘catch-state’ that some biological muscles such as mollusks have. Catch-state or lock-state is when the muscle locks in its current length without consuming energy. We have observed a linear correlation between the annealing temperature and the ratio of the locked amplitude/force to the active amplitude/force (catch-index, hereafter) (Figure 4A). This catch-state happens when we increase the temperature of the muscle to a point higher than the temperature at which the muscle was annealed. So in the long run over multiple cycles during the actuation the muscle will anneal, which reduces the catch-index. This problem of temporary catch-state can be resolved by designing a locking mechanism. But if, for applications such as active catheters, having a catch-state for only several cycles is desirable, the cost of nylon is cheap enough to avoid adding any accessories to obtain a permanent catch-state. As Figure 4B indicates, the actuator holds around 160 mN out of the 1400 mN peak force it generated in the active state.

By roller-pressing the nylon through the V grooves of the rolling mill we created nylon square rods. Following the same

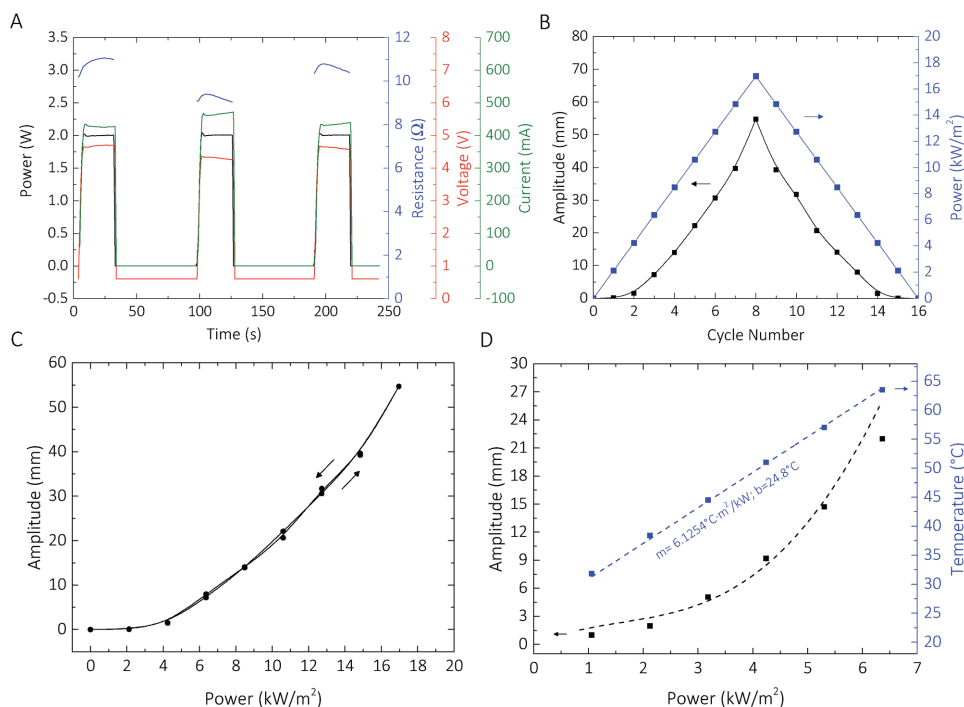


Figure 3. A) Performance of the constant power source setup. The black line represents the power as a function of time. The convergence is less than 3 s to the target power (in this case 2 W). The resistance during the actuation changes by almost 1 Ω (blue line) which justifies the need for the constant power source. B,C) Amplitude of a bending actuator for a symmetric power cycling. The plots show a very symmetric and reversible amplitude response. (The unit kW m^{-2} is the same as mW mm^{-2} .) D) Amplitude and temperature of a bending actuator as a function of input power in the form of continuous cycling for each power value. The blue dashed line is a linear fit and the black dashed line is what our model predicts for this experiment.

fabrication procedure for the bending actuator, we created four Joule heating traces with our conductive paint (Figure 1D). When power is applied to each side of the actuator, we can achieve bending in $\pm X$, $\pm Y$ and even $-Z$ direction if all of the four sides of the rod are excited at the same time. To provide each side of the actuator with variable voltages we used a half-bridge rectifier made of Schottky diodes (Figure S6, Supporting Information). By applying two sinusoidal signals 90° out of phase to the opposite sides of the actuator (top and bottom connected to $V_0 \sin(\omega \cdot t)$, left and right connected to $(V_0 \sin(\omega \cdot t + \pi/2))$) the actuator's tip traversed a circular path (Figure 4D, Video S2, Supporting Information). Similarly, by applying two triangle signals with a phase difference of 90° , a square path was traversed by the actuator's tip (Figure 4E, Video S3, Supporting Information). When two sinusoidal waves with one having double the frequency of the other one and 90° out of phase were applied, more complex trajectories such as Lissajous figures were observed (Figure 4F, Video S4, Supporting Information).

We measured the frequency response of the bending actuators with two different dimensions (Figure 4G, Video S5, Supporting Information). The thinner actuator had resonance frequency of 8.96 Hz while the other showed resonance frequency of 16.78 Hz. From the equation for the natural frequency of cantilever beams^[26] we can write:

$$\omega_n \sim \frac{t}{L^2} \sqrt{\frac{E}{\rho}} \quad (3)$$

where t is the thickness of the actuator, L is the length of the beam, E is Young's modulus, and ρ is the volumetric mass density. The ratio of the resonance frequency of the beams matches with the ratio of their t/L^2 which shows that we can control the resonance frequency by just adjusting the dimensions.

We used two 6 W blue (450 nm) laser diodes to excite a thin nylon beam (l, w, t : 21.5 mm, 0.7 mm, 112 μm) coated with thermally conductive paint (Figure S3, Supporting Information). Since the cooling time constant scales linearly with the thickness, by making the beams thinner we can expect faster cooling time and therefore, higher excitation rates. The conduction across the thickness of the nylon scales with the square of the thickness, which means the amplitude reaches steady state faster. By exciting the beam with laser diodes at an optical output power of 65 mW the peak-to-peak amplitude of 2.5 mm at 2 Hz was measured. Since the laser beam is only heating a very small area on the surface of the beam, the displacement over length is smaller than what we observed from the Joule-heated actuators (Video S6, Supporting Information).

We measured the performance of a nylon bending actuator (l, w, t : 90 mm, 3 mm, 0.87 mm) at input power of 8.15 kW m^{-2} . The actuator showed less than 5% deterioration in the amplitude over 100 000 cycles (Figure 4H). This relatively long cycle life enables this technology to be used in many applications such as tactile displays, active catheters, dynamic sculptures, toys, optical shutters, and many others.

To effectively stimulate the mechanoreceptors in order to give us a sense of touch, a minimum force of 30–50 mN is

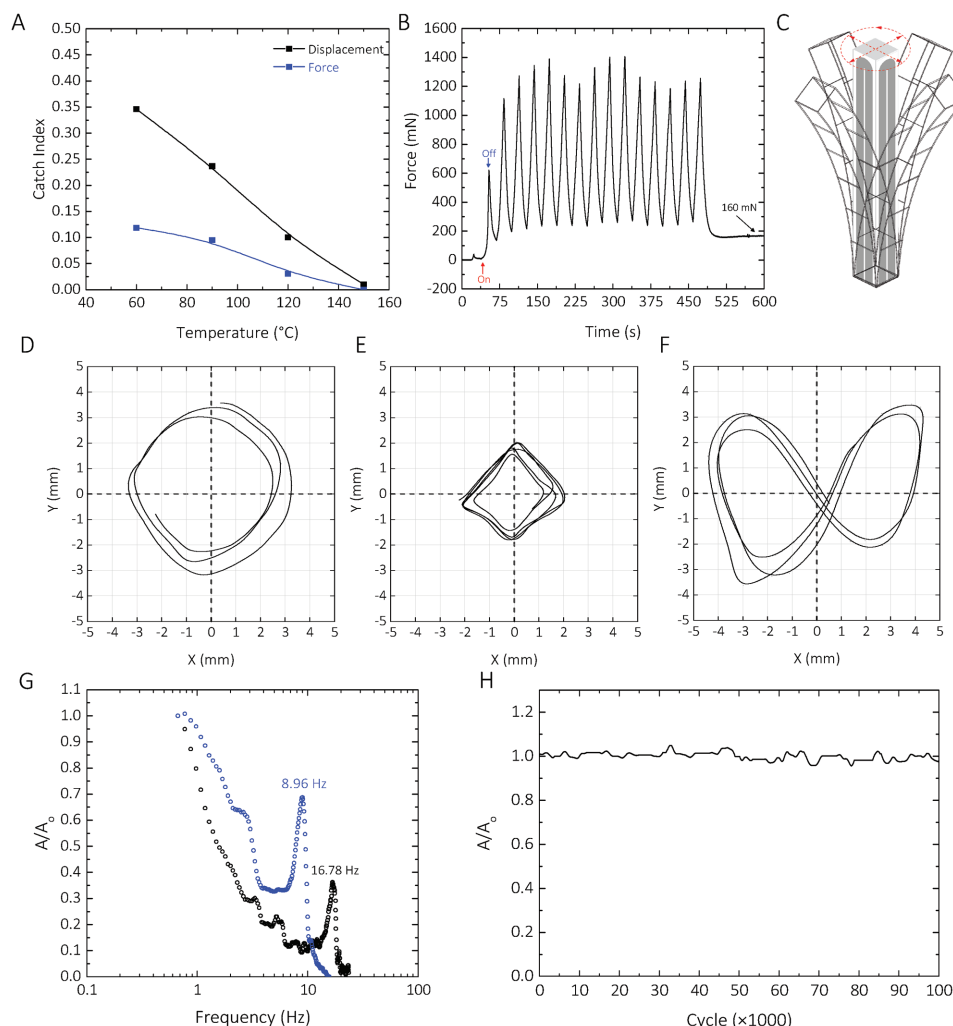


Figure 4. A) Catch-index as a function of the annealing temperature shows an almost linear relationship for both the displacement and force. B) A nylon beam (l,w,t: 66 mm, 2.65 mm, 0.873 mm) annealed at 60 °C shows a catch-index of around 0.12. C) Schematic of the multidirectional actuator. D–F) The coordinate of the tip of the actuator by applying the following inputs (left to right): X-Ch: $V_x = V_{x0} \sin(\omega t)$, Y-Ch: $V_y = V_{y0} \cos(\omega t)$; X-Ch: $V_x = V_{x0} \text{triangle}(\omega t)$, Y-Ch: $V_y = V_{y0} \text{triangle}(\omega t + \pi/2)$; X-Ch: $V_x = V_{x0} \sin(2\omega t)$, Y-Ch: $V_y = V_{y0} \cos(\omega t)$. G) Frequency response of two nylon bending actuators with dimensions of l,w,t: 60 mm, 1.54 mm, 0.322 mm (represented by blue circles) and 72 mm, 2.3 mm, 0.85 mm (represented by black circles) shows resonance frequencies of 8.96 and 16.78 Hz, respectively. H) Life cycle of a bending actuator (l,w,t: 90 mm, 3 mm, 0.87 mm) shows less than 5% deterioration in amplitude response over 100 000 cycles in a non-temperature controlled environment.

required.^[27] The force generated by nylon bending actuators satisfies this requirement. In addition to this, the relatively good response time of the actuator makes it a potential candidate for use in the design of a tactile display. Due to the geometry and functionality of the nylon multidirectional actuator, it can possibly be used to navigate through blood vessels or other internal organs. The key requirements for navigating through blood vessels are small diameter (<1.65 mm), a high degree of bending (>20°), a high enough force (≈25 mN at the tip), and adequate response time (on the order of seconds),^[28] all of which these nylon bending actuators satisfy.

By exploring and using the intrinsic properties of highly oriented nylon filaments we proposed a new design for bending artificial muscles. It has already been shown that by twist-coiling highly oriented nylon filaments we can amplify the tensile stress (up to 49%), here we are showing

that by making nylon beams from highly oriented nylon filaments (such as high performance fishing lines and sewing threads) we can achieve a displacement-to-length ratio of 125%, but with a much lower output force. This combination of displacement and output force has applications in many devices such as surgical tools, toys, and smart windows, to name just a few. We believe that this new type of actuator will create an avenue for new artificial muscle designs made from nylon.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author. The supporting information includes materials and methods, supporting text, figures, tables, and videos.

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