

Multi-Stimulus Soft Actuators from Aerosol Jet Printed MXene-Cellulose Composite

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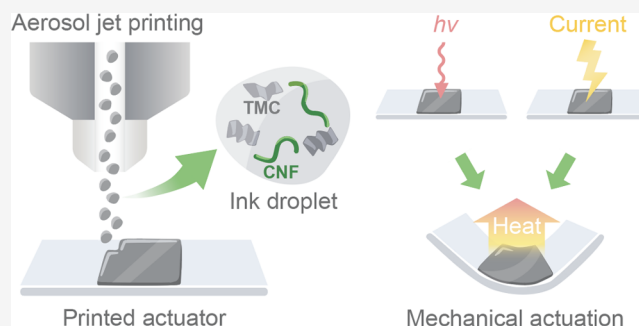
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ABSTRACT: The evolution of next-generation flexible, stretchable, and conformal microrobotics hinges significantly on advances in fabrication methods. There is a need to enhance the scalability of actuator feature dimensions and to increase the design versatility. This study demonstrates the additive manufacturing of soft multi-stimulus thermomechanical actuators based on a composite of transition metal carbide (TMC, MXene) and cellulose nanofiber (CNF) nanomaterials using aerosol jet printing (AJP) for scalable/customizable actuation. Aqueous MXene-CNF films with tunable electrothermal and photothermal properties were printed onto polycarbonate (PC) membranes for wired and/or wireless operation, having full reversibility with induced heating below 120 °C. Printed devices as small as 5 mm² and 2.5 mm² yielded sharp curvatures of 0.92 cm⁻¹ and 0.87 cm⁻¹, respectively. By printing MXene-CNF to select regions on both sides of the substrate, complex actuation modes were realized with directional, reversible control. This work highlights the promise of AJP-fabricated MXene-CNF systems for soft robotics applications.

KEYWORDS: MXene, Soft Robotics, Thermomechanical Actuation, Electromechanical Actuator, Photomechanical Actuator, MXene-CNF



Actuators convert input energy (electrical,¹ thermal,² electro-magnetic,³ etc.) into mechanical energy. Thermomechanical actuation is driven by alteration of moisture content,⁴ thermal expansion,⁵ and/or changes in physical properties.⁶ Soft actuators are composed of human-friendly materials such as polymers and hydrogels for bending,⁷ stretching,⁸ and surface conformity.⁹ These soft materials enable adaptability,¹⁰ safety,¹¹ and fragile object manipulation,¹² leading to soft robotics in the form of artificial muscles,¹³ switches,¹⁴ micro-electromechanical systems (MEMs),¹⁵ and wearables.¹⁶

Battery-free operation of soft actuators reduces weight, size, and cost while increasing mobility, payload, and sustainability. Wireless control is possible by illuminating near-infrared (NIR) light onto transition metal carbide films (TMC, Ti₃C₂T_x, a 2D material in the MXene family).¹⁷ Notable achievements with soft actuators from MXene include the self-folding box, self-assembling flower, traveling worm, and multimodal snake.^{17–21} Furthermore, the actuation response of MXene-based actuators can be enhanced through integration of cellulose nanofiber (CNF)¹⁷ or through surface treatments, which have been shown to yield a nearly 30% bend angle increase.²² Despite encouraging progress, reports on the miniaturization of wireless soft actuation systems are limited, in part due to fabrication challenges.¹⁷

Soft robotics fabrication methods include electrospinning,²³ knitting,²⁴ solution casting,²⁵ spray coating,²⁶ and weaving.^{24,27}

Additive manufacturing, including 3D printing,²⁸ inkjet printing,²⁹ and aerosol jet printing (AJP),^{30–32} enables rapid mask-free prototyping of custom actuator designs. Thin films can be fabricated by AJP in a single step at high resolution,³³ low cost,^{30,34,35} and room temperature onto chemical/heat-sensitive, flexible, and/or curved substrates.^{34,36} Having a 10 μm resolution limit, AJP enables the printing of microscale soft actuators.

In this work, MXene-CNF actuators are fabricated by AJP. MXene-CNF coatings were printed onto flexible PC membranes for soft robotics applications, demonstrating microscale features, directional bending control, and a sleeping flower (Figure S1). Actuators respond to both electrical and photonic stimuli for wired or wireless operation. Fully printed actuators exhibited a linear power–bend relationship, with full recovery after stimulation at low temperatures for over 1000 cycles.

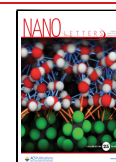
Given prior success, PC membranes were selected as soft substrates.^{37–39} Transition metal carbide (TMC, MXene) was

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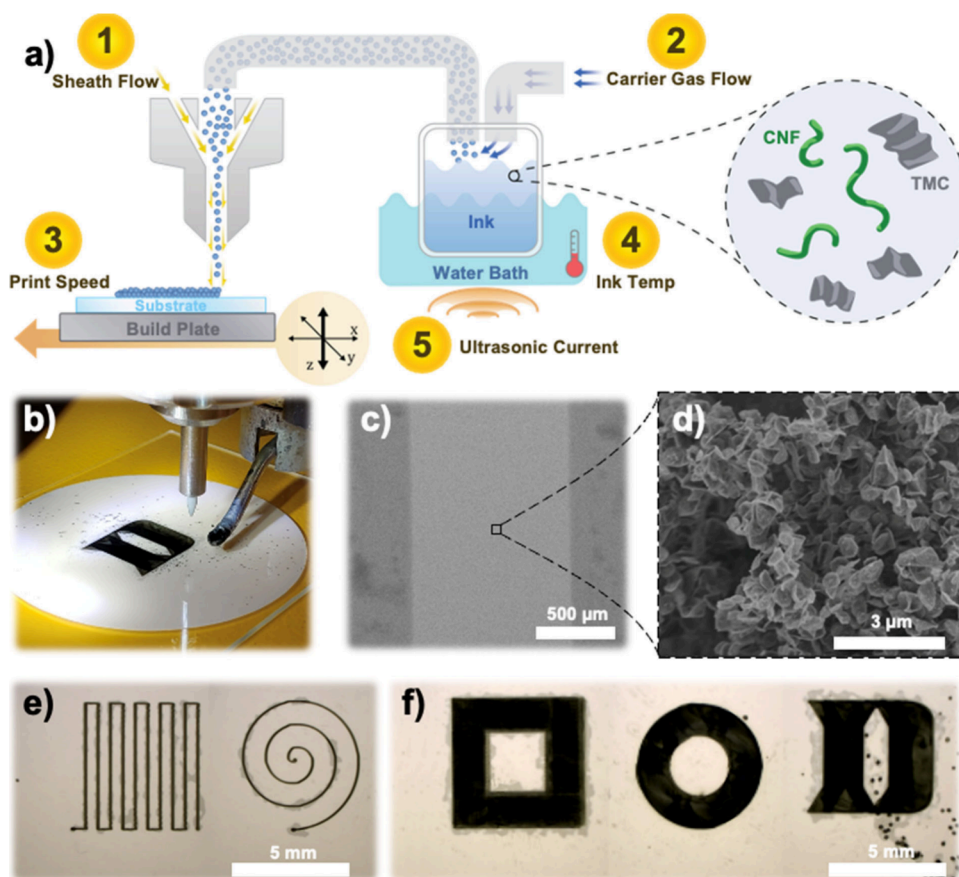


Figure 1. Aerosol jet printing of MXene-CNF films. (a) Five key process parameters are controlled for governing quality and dimensions of MXene-CNF films printed at room temperature, including sheath flow, carrier gas flow, print speed, ink temperature, and ultrasonic current. (b) Direct writing of a MXene-CNF-based coating in the pattern of a Duke University logo. (c, d) Scanning electron microscopy image of 1 mm-width printed MXene-CNF thin film on silicon. (e) Single trace print patterns demonstrate capability for drawing straight and curved single traces. (f) Multiple adjacent printed traces enable drawing of complex shapes having 90° corners, curved features, and sharp angles.

chosen due to its exceptional photothermal characteristics. Aqueous MXene inks were formulated with cellulose nanofibers (CNFs), for actuation enhancement.¹⁷

Print deposition and performance are governed primarily by AJP process control parameters including carrier gas flow (N_2 , SCCM), sheath gas flow (N_2 , SCCM), ultrasonic atomizer current (mA), and print speed (mm/s) as illustrated in Figure 1a.⁴⁰ Print process optimization enabled printing for long print times without clogging, enabling large features (Figure 1b–d). Ink splatter can be reduced with integration of a divert/boost system.⁴¹ Print optimization on glass mitigated undesirable phenomena, such as the coffee ring effect. Further print optimization on PC improved print quality in terms of edge definition and reduced width variations (Figure 1e). Printing multiple adjacent single traces gives rise to filled features having straight or curved traces with sharp angles (Figure 1f).

A printing process was developed for stabilizing deposition, dimensional control, and resistance tuning of MXene-CNF films. Printing at room temperature is often challenging due to the buildup of solvent.³⁴ Optimization of print parameters enabled printing at sub-millimeter resolution. While glass is an inexpensive and convenient substrate for print prototyping, a substantial portion of prototyping occurred on porous polycarbonate (PC) membranes. When printing aqueous inks, water buildup and subsequent runny printed films, evidence of spillage, can be reduced by increasing the hydrophilicity of the substrate (water on glass having a static

contact angle of 47–58° while water on rigid PC having a static contact angle of at least 85°).^{42–44} However, faster deposition rates were possible on PC membranes without observing runny printing because porous PC exhibits excellent water retention due to the ink soaking into the pores. This enhances the print fidelity and visual quality of printed films relative to glass (see Figure S2). Printing multiple devices of the same design and identical process control parameters gives rise to a 7× conductance increase in MXene-CNF films on PC as compared with glass (Figure 2a–c). Excess fluid accumulation and uncontrolled spreading during printing limit the effective material deposition along the electron transport pathway, which accounts for the 7-fold increase in conductance. Increasing the print speed further prevents film thickness variations and circuit shorting due to runny films (Figure 2d). Runny printing is reduced by decreasing the atomizer flow and sheath flow (see Figure S3). Increased film thickness is realized by either increasing atomizer flow or increasing the number of print passes, consistent with other reports (Figure 2e,f).^{34,40,45} Conductance increases with film thickness due to the larger network of conductive paths.

When Joule heating drives actuation, the electronic resistance dictates voltage operation requirements. Precise control of print speed and measuring resistance revealed a linear speed–resistance dependence enabling resistance tuning of MXene-CNF films (Figure 2g). Compared to silver nanowire (AgNW) films, among the best for sinter-free

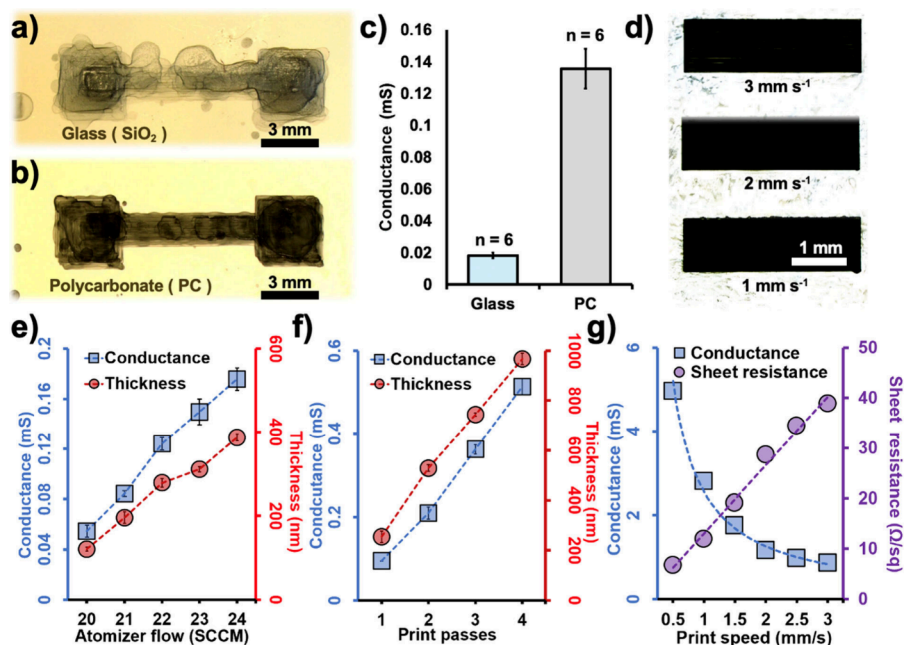


Figure 2. MXene-CNF print process control. (a) Optical microscopy of MXene-CNF electrodes printed onto nonporous glass as compared with a porous and water-absorbing (b) polycarbonate membrane at the same print conditions. (c) Bar graph indicating relative conductance of films on different substrates (sample size of six devices and error bars represent standard intervals). (d) Increased print speed results in thinner films with improved visual quality. (e) Conductance and thickness of printed MXene-CNF-based thin films on glass varying the atomizer flow rate and (f) number of print passes at an atomizer flow rate of 25 SCCM. (g) Speed and resistance have linear dependence, enabling resistance control when printing MXene-CNF films on PC.

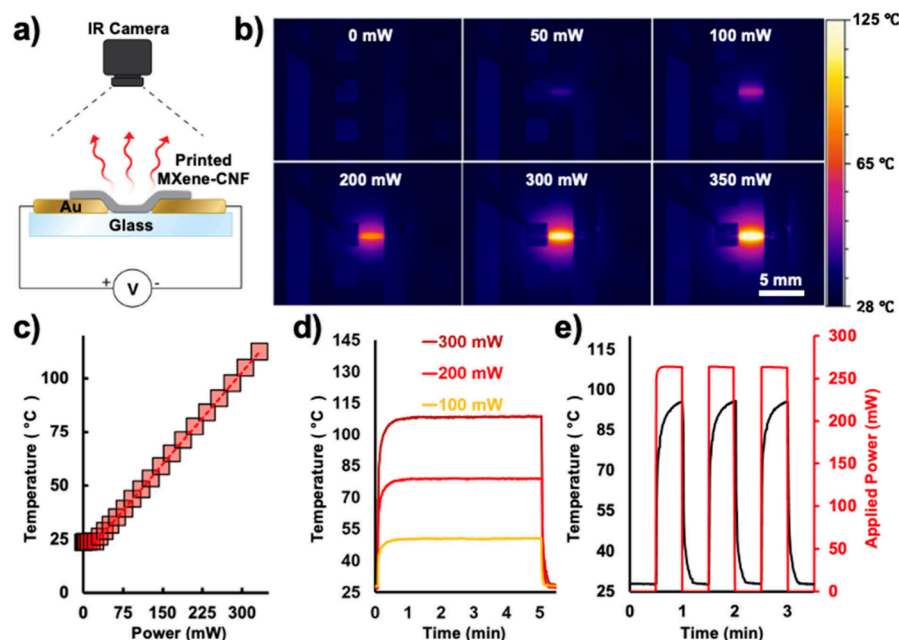


Figure 3. Thermal performance of printed MXene-CNF microheaters. (a) Side-view schematic illustration of experimental setup for measuring electronically induced temperature response and relaxation time of printed MXene-CNF microheaters. (b) Thermal map images showing the top-down view of a representative microheater (100 μm × 3 mm) on glass under different applied power conditions. The resistance of the microheater is 400 Ω, corresponding to a sheet resistance of approximately 13.3 Ω/sq. (c) Electrode peak temperature of the MXene-CNF microheater exhibiting linear dependence on applied electrical power. (d) Temperature versus time, showing stability under constant power for a duration of 5 min. (e) Voltage applied in cycles demonstrates consistent heating performance of a microheater (100 μm × 3 mm) when applying 10 V pulses (approximately 263 mW) for 30 s duration over three cycles.

conductivity, MXene-CNF films exhibited $\sim 30\times$ less conductance (see Figure S4a), given the same print path and print duration. Increasing print duration reduces resistances; however, a reduced speed increases risk for runny prints due

to solvent buildup, causing deviation in the speed–resistance linear fit (see Figure S4b). Therefore, when printing thick films with an aqueous MXene-CNF ink at room temperature, especially if a low resistance is desired, it is recommended to

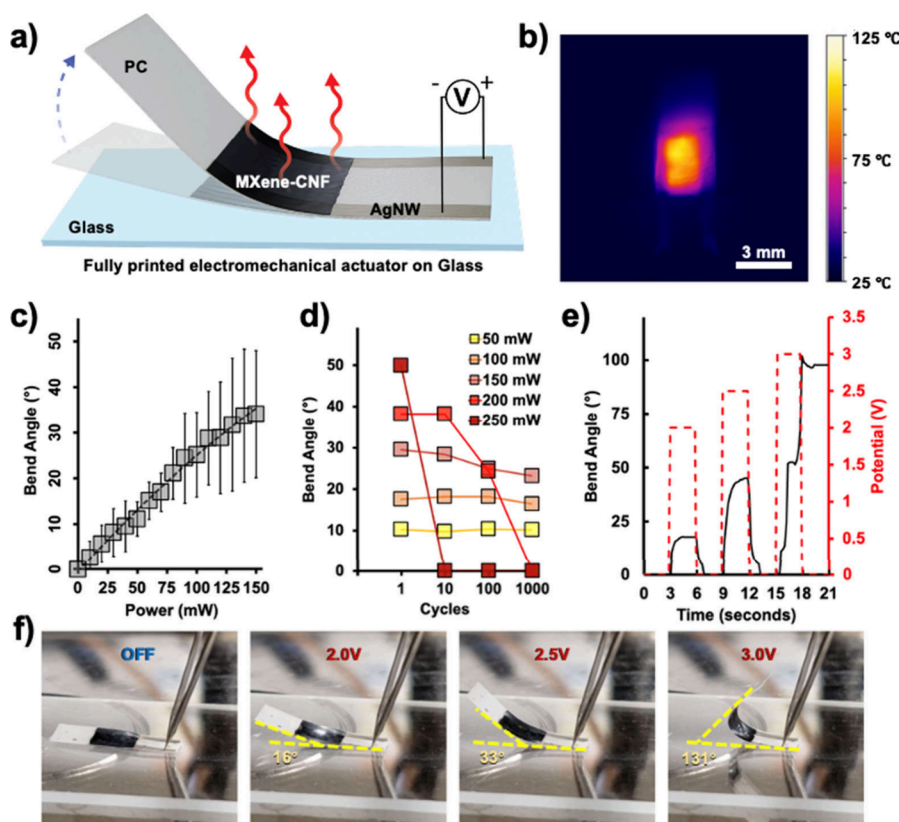


Figure 4. Electronic actuation of printed multi-stimulus actuators. (a) Side-view graphic of multi-stimulus actuator having printed a 3 mm $\times$ 4 mm MXene-CNF resistor bridging two printed 500 μ m $\times$ 8 mm AgNW electrodes on a 3 mm $\times$ 12 mm PC substrate resting on a glass slide. (b) Top-down heat map image captured while applying 100 mW to a multi-stimulus actuator clamped on both ends to maintain camera focus. (c) Maximum bend angle achieved versus applied power up to 150 mW for the device in the absence of a substrate, when 2/3 (8 mm) of the device was suspended in air while the other 1/3 (4 mm) rested on a glass substrate (sample size of three devices). (d) Device cycling endurance in the absence of a substrate. (e) Time required to achieve maximum bend angle for a representative device resting on glass. An applied potential of 2 V ($\sim$ 36 mW), 2.5 V ($\sim$ 65 mW), and 3 V ($\sim$ 111 mW). (f) Photo captured at maximum bend angle while applying voltage ranging from 0 to 3 V to a representative multi-stimulus actuator resting on glass.

print multiple passes, pausing between passes to ensure sufficient time for evaporation. Like AgNWs, aerosol jet printing MXene-CNF films is simple, fast, and cost-effective, as the films are functional immediately after printing without requiring any postprocessing steps such as vacuum drying, thermal annealing, and/or chemical treatment, each adding complexity, time, and costs.

Actuation is primarily driven by the generation and localization of thermal energy and mismatch in thermal expansion coefficients in the bimorph.^{17,46,47} Electrical stimulation was provided for evaluating the thermal performance of heating element prototypes with sub-millimeter film widths, herein referred to as microheaters. Thermal performance of actuators can be difficult to assess because, without clamping methods, the device can move out of the infrared-camera focus. Therefore, thermal performance characteristics for printed MXene-CNF coatings were studied on glass by capturing 2D thermal maps and by investigating temperature modulation and heat stability.

A schematic detailing the configuration of the printed MXene-CNF microheaters is shown in Figure 3a. Preformed gold contact pads were used for interfacing with the printed film, which was printed to bridge the pads. The soft gold pads provide good interfacing with a signal modulation unit, and the low resistance of the gold ensures Joule heating occurs predominantly in the relatively higher resistance region of the

printed MXene-CNF bridge. Thermal 2D heat map images for a representative MXene-CNF microheater are shown in Figure 3b. Localization of the heating is evident from the heat map images, particularly at higher applied power, where the microheater reached peak temperatures above 100 °C.

A linear dependence of peak temperature and applied power was observed for the printed microheaters at >50 mW (Figure 3c). This indicates that a desired target temperature can be achieved for a given application within the recorded temperature range. However, the device may not be used as a resistive thermal detector, given that resistance and temperature were nonlinear, suggesting an altering of material structure such as thermal expansion, microfractures, or changes in interlayer spacing due to loss of moisture content as reported by Sun et al. in 2019 (see Figure S5).⁴⁸ While maintaining a constant potential (voltage) is generally sufficient for temperature control, variations in resistance, given their influence on the voltage–temperature and current–temperature relationships, can lead to reduced precision. Therefore, applying constant power (in milliwatts) offers improved accuracy in temperature regulation. A constant applied power enables a maximum temperature to be realized within 60 s, and the temperature, even if above 100 °C, is stable for several minutes (Figure 3d). A temperature above 105 °C is achieved at a constant power supply equal to 300 mW, corresponding to a power density of 1 W/mm². To control power, the user must have additional

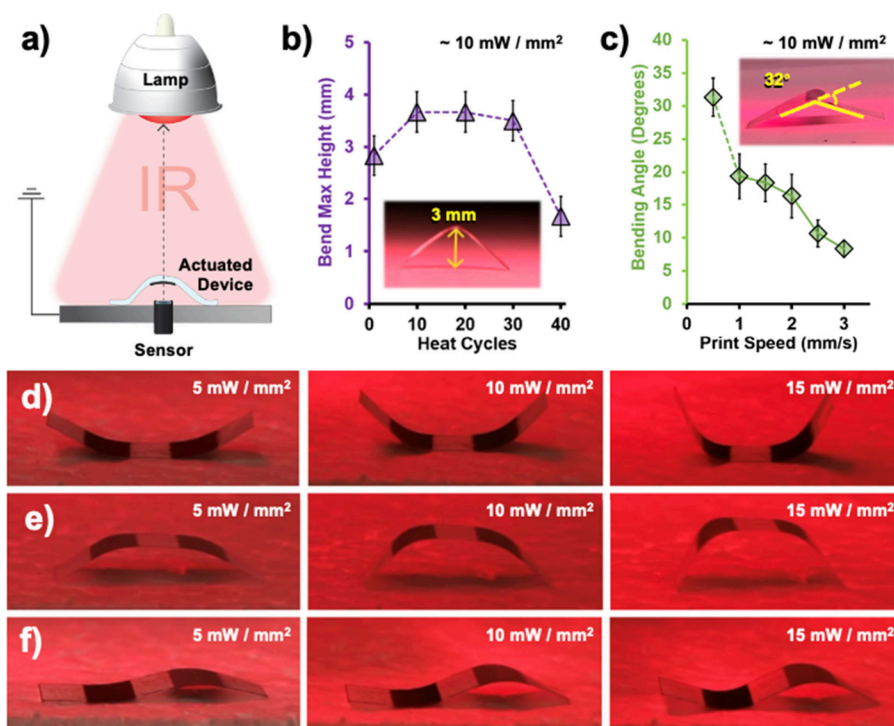


Figure 5. Wireless control of printed soft actuators via infrared radiation. (a) Side-view graphic detailing experimental setup for remote actuation. (b) 3 mm $\times$ 5 mm printed MXene-CNF actuator on PC and resulting heat cycle endurance based on the maximum height while bent (sample size of three devices and error bars representing standard error intervals) when supplying an NIR power density of approximately 10 mW/mm². (c) Bending angles of 3 mm $\times$ 10 mm actuators printed at speeds ranging from 0.5 to 3 mm/s showing the impact of print speed (and thus MXene-CNF film thickness) on resultant actuation (sample size of three devices and error bars representing standard error intervals) when supplying an NIR power density of approximately 10 mW/mm². Pictures showing thermomechanical response at different power densities of a 4 mm $\times$ 20 mm actuator having two printed 4 mm $\times$ 5 mm coatings on the (d) same side, (e) same side reversed, and (f) opposite sides.

components to measure and manually or programmatically adjust voltage in real-time, thus complicating and adding cost to thermal control. The ability to set voltage and achieve a desired temperature is demonstrated with the application of a square-waveform potential–time profile, and reproducible heating behavior was observed after multiple successive 30 s cycles (Figure 3e). Under an applied 10 V ($\sim$ 263 mW), the microheater reached 70 °C, 80 °C, and a maximum temperature of 95 °C within 1, 4, and 23 s. The film cooled when the voltage was switch off and returned from 95 °C to within one degree of ambient temperature ($\sim$ 27.5 °C) in 12 s on average.

As previously noted, the MXene-CNF microheaters serve as a thermomechanical actuation layer when printed onto flexible PC substrates. With the thermal performance of the printed films characterized, electrically controlled soft actuators were next studied. In these soft actuators, control of temperature is essential, as it regulates the moisture content, which dictates the extent of actuation. A representative graphic for the device design is shown in Figure 4a and Figure S6, which illustrate the film actuating the PC under applied electrical potential. This fully printed design features a printed MXene-CNF microheater bridging two printed AgNW leads on a PC membrane. AgNWs were selected for their significantly lower resistance (compared to the MXene-CNF) to ensure that the majority of power loss and thus Joule heating occur at the MXene-CNF coating when applying an electrical stimulus.

A power of 100 mW was applied to a representative device resting on a glass slide, and the resulting heat map is shown in Figure 4b. The actuator was clamped on both sides to restrict

bending. Note, the heat map appears to show less uniform heat distribution due to distortion from slight mechanical actuation of the film, which could not be prevented. To actuate, the bending force must overcome the adhesion force at the interfacial surface between the hydrophilic PC and the underlying substrate. Glass and PC adhere well, as both surfaces are hydrophilic. Less applied power was needed for equivalent bending when the device rested on a polystyrene (PS) substrate due to less attraction between hydrophilic PC and hydrophobic PS. At a further extreme, even less power was required in the absence of a substrate, when 2/3 (8 mm) of the device was suspended in air while the other 1/3 (4 mm) rested on a glass substrate, further illustrating the contributing role of substrate adhesion forces in achieving actuation of the PC. The actuation power threshold, defined as the minimum amount of power required to cause movement that was detected by the unaided human eye, is shown for each case (see Figure S6). The adhesion force at the interface is proportional to the effective contact area, which can vary substantially between devices due to slight differences in how each device settles on the surface. This variability accounts for the large experimental variation observed, as reflected by the standard error bars in the corresponding bar plot.

The average bend angle increases linearly with power and temperature (Figure 4c), consistent with a recent report involving MXene-CNF on PDMS.⁴⁹ The notable increase in variability at a power above 75 mW is attributed to differences in contact surface area in the area where half of the substrate is resting on glass due to some extent of actuation and relaxation occurring at the PC–glass interface. Actuators are able to

survive over 1000 cycles with full recovery (see Figure S7) at low temperatures (Figure 4d). Actuation under constant applied voltage resulted in a maximum bending angle in 2.08 s at 2 V (~ 36 mW), fully relaxing in 1.25 s and achieving a maximum bend angle in 2.98 s at 2.5 V (~ 65 mW) and fully relaxing in 1.5 s (Figure 4e). Differences in power–bending relationship are attributed to the thickness of the MXene-CNF film. Thicker films have faster cooling rates and therefore require higher power to achieve the same temperature. The actuation was found to be reversible for both the 2 and 2.5 V scenarios. Applying more voltage increased the bending, with 3 V (corresponding to ~ 110 mW) yielding a 131° angle or curvature (2.86 cm^{-1}); however, the actuation was not reversible at this higher voltage. Increasing power and multiple thermal cycles can damage the film by generating regions with high electrical resistance. Irreversible actuation is attributed to the softening and stiffening of the PC substrate about the glassy transition temperature, T_g , measured by means of flash differential scanning calorimetry by Shamim, Nabila, et al. and reported as $120\text{--}138^\circ\text{C}$.⁵⁰ Furthermore, drying of cellulose and associated water loss can lead to an irreversible aggregation, commonly described as hornification.⁵¹ Initially, when applying a power of 100 mW to a device of the same design, the heating is uniform and the peak temperature is around $105\text{--}110^\circ\text{C}$. Then, the same device was damaged due to an increased applied power (140 mW), leading to temperatures above 200°C . When returning to an applied power of 100 mW, temperatures above 145°C were observed due to localized defects leading to nonuniform heating (see Figure S8).

The printed MXene-CNF film has been shown to yield controllable heating and actuation under electrical stimulus, but this requires a wired connection to the device. For wireless actuation, the use of NIR irradiation was also examined. The experimental setup used to quantify wireless actuation driven by NIR exposure at different power densities and durations is illustrated in Figure 5a. Reversibility and operational durability were evaluated through cyclic actuation testing to failure, wherein $3\text{ mm} \times 5\text{ mm}$ printed actuators were repeatedly exposed to NIR illumination ($\sim 10\text{ mW/mm}^2$) sufficient to induce consistent bending angles exceeding 90° or curvature (1.21 cm^{-1}) across more than 30 cycles (Figure 5b). Each actuation cycle consisted of a controlled irradiation phase, during which the actuator reached its maximum deflection, confirmed by allowing approximately 5 s after the motion ceased, followed by a thermal relaxation phase lasting 10 s after the NIR source was turned off. A positive correlation between film thickness (thicker films were realized at lower printing speeds) and achievable bend angle was observed, indicating that increased photothermal mass contributes to greater mechanical deformation (Figure 5c).

To explore the performance of thermomechanical actuation under photonic stimulus at reduced dimensions, miniaturized actuators with microscale printed features were fabricated and characterized. Designs included a grid pattern, a vertical stripe, and horizontal stripe patterns. For each device, bending was observed, although the threshold irradiance required to initiate actuation of devices having $1\text{ mm} \times 5\text{ mm}$ patterns was found to increase with reduced MXene-CNF surface coverage, given more heating overall and therefore more total water loss (see Figure S9). The actuation was not completely reversible for any of the $1\text{ mm} \times 5\text{ mm}$ patterns, likely due to the higher

temperatures required to achieve sufficient water loss for actuation.

A key advantage of aerosol-jet-printed MXene-CNF films on polycarbonate (PC) substrates is the ability to impart directional control by selectively patterning coatings on one or both sides of the actuator. Regardless of the angle of incident near-infrared (NIR) illumination, the actuators consistently bend toward the side bearing the MXene-CNF film. This behavior is fully consistent with the bimorph mechanism, which originates from the mismatch in thermal expansion coefficients between PC and the MXene-CNF composite. Upon NIR heating, the PC undergoes greater thermal expansion, relative to the coating. Because the two layers are constrained together, this differential strain generates interfacial stress: the PC layer experiences compressive stress near the interface, while the MXene-CNF layer resists expansion, creating a net bending moment that deflects the actuator toward the coated side. Thus, lifting of the edges occurs when the coating and light source are on the same side, whereas lifting at the center of the actuator is observed when illumination is applied to the uncoated side (Figure 5e,f). Furthermore, printing coatings on opposite sides of the PC membrane enables differential actuation across the length of the device, producing complex deformation profiles such as sinusoidal bending patterns (Figure 5g).

As previously noted, thermomechanical actuation under electrical stimulus was realized with the fully printed design featuring a printed MXene-CNF microheater bridging two printed silver nanowire (AgNW) leads on a PC membrane. With this design, the device was postulated to be responsive to both electronic and NIR powering methods for wired and wireless actuation. AgNWs were selected as leads, given superior electronic conductivity and a lower photothermal conversion efficiency. Furthermore, the MXene-CNF film coverage area was much larger than the AgNW coverage area to ensure that the majority of heating happens in the target region. Actuation was realized when illuminating the multi-stimulus device to 10 mW/mm^2 , 20 mW/mm^2 , and 50 mW/mm^2 power densities (see Figure S10).

This work provides a foundation for room-temperature fabrication of fully printed, multi-stimulus soft actuators. Using AJP technology, water-based conductive inks were printed onto lightweight, porous, and flexible polycarbonate membranes. The actuators were functional immediately after printing, without requiring any postprocessing steps such as vacuum drying, thermal annealing, and/or chemical treatment, which can be complex, time-consuming, and/or expensive. Also, by modifying the print path design and optimizing the printing process parameters, control over the film patterning, down to the sub-millimeter scale, was demonstrated in terms of shape and dimensions, including width, length, and thickness. Thermomechanical soft actuators were demonstrated to work under an electrical or photonic stimulus with reversible bending behavior. Actuators can survive up to 30° bend angles for over 1000 cycles. While controlled with a photonic stimulus using NIR light, the soft actuators demonstrated reliable bending, directional control, and wireless responsiveness. Cycling performance can be improved at lower temperatures or by substituting PC for a substrate that can handle higher temperatures. Importantly, filled 1 mm -width devices yielded sharp angles as significant as 132° or curvature (0.92 cm^{-1}), while partially filled 1 mm -width devices yielded as much as 125° angles or curvature (0.87

cm⁻¹), showing potential for scalable manufacturing. The versatility of the AJP process was demonstrated through selective patterning of the MXene-CNF coating on both sides of the substrate, enabling directional, reversible, and complex actuation modes. In the future, AJP of MXene-CNF onto more heat resilient substrates may be a viable route for advancing lightweight, battery-free soft actuators. Altogether, this work establishes a scalable and versatile fabrication strategy that advances the integration of MXene-CNF-based soft actuators into next-generation miniaturized robotic systems, wearable devices, and programmable multifunctional platforms.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.nanolett.5c03256>.

Experimental sections, substrate preparation, ink formulation, aerosol jet printing, electrical characterization, optical characterization, thermal characterization, supplemental figures, and additional notes (PDF)

Movie S1: Reversible actuation under electrical stimulus (MP4)

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Author Contributions

B.C. and A.F. designed this project. B.C., A.B., J.H., T.H., and M.B. operated the aerosol jet printer. B.C. designed sputter masks and printer files. B.C. and W.C. designed and supported graphics. B.C., A.B., J.H., T.H., and M.B. performed experiments related to multi-stimulus performance of printed actuators. B.C. and A.F. conducted analysis related to optical, electrical, and thermal characterization as well as actuation performance. B.C. and Y.Z. performed experiments assessing the performance of multi-stimulus actuators. B.C. and A.F. wrote the manuscript. B.C., W.C., and A.F. helped prepare the figure set. H.W. and A.F. supervised the project and edited the manuscript. All authors approved of the manuscript for submission.

Notes

The authors declare no competing financial interest.

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■ ABBREVIATIONS

TMC, transition metal carbide; MXene; CNF, cellulose nanofibril; PC, polycarbonate; AgNW, silver nanowire

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