High-Performance, Low-Voltage, and Easy-Operable Bending Actuator Based on Aligned Carbon Nanotube/Polymer Composites

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ABSTRACT In this work, we show that embedding super-aligned carbon nanotube sheets into a polymer matrix (polydimethylsiloxane) can remarkably reduce the coefficient of thermal expansion of the polymer matrix by two orders of magnitude. Based on this unique phenomenon, we fabricated a new kind of bending actuator through a two-step method. The actuator is easily operable and can generate an exceptionally large bending actuation with controllable motion at very low driving DC voltages (<700 V/m). Furthermore, the actuator can be operated without electrolytes in the air, which is superior to conventional carbon nanotube actuators. Proposed electrothermal mechanism was discussed and confirmed by our experimental results. The exceptional bending actuation performance together with easy fabrication, low-voltage, and controllable motion demonstrates the potential ability of using this kind of actuator in various applicable areas, such as artificial muscles, microrobotics, microsensors, microtransducers, micromanipulation, microcantilever for medical applications, and so on.

KEYWORDS: carbon nanotube · polymer · composite · actuator · bending · thermal

Materials that can directly convert different types of energy into mechanical energy are commonly known as actuator materials. They are expected to be widely used in many applications, such as artificial muscles, robotics, optical switches, micropumps, microsensors, and so on.[1−6] High-performance and lightweight actuator materials are in urgent need to replace conventional electromagnetic motors and combustion engines, which suffer from the drawbacks of cumbersomeness, inflexibility, and low efficiency.[5,6] In the past few decades, electroactive polymer actuators have been widely investigated thanks to their properties of lightweight, flexibility, and low cost. They can be broadly divided into two categories based on their mechanism of actuation: ionic and field-activated.[6] Among the ionic classification, the actuators were mostly actuated in an electrochemical system using a liquid electrolyte.[1,7−10] Some of them are based on carbon nanotubes (CNTs), for CNTs have unique electrical, thermal, and mechanical properties (*e.g.*, excellent electrical and thermal conductivity and high mechanical strength).[1,7−9] However, it is noted that the actuations mostly occur only in electrolyte solutions, which are inconvenient for practical application, because of the requirement of high-standard safety encapsulation of the liquid electrolyte. In the field-activated classification, dielectric elastomer actuators are attractive due to the advantages, such as large strain, high speed of response, high energy densities, and so on.[2,3,6,11] However, owing to their low dielectric constants relative to ceramics, dielectric elastomers typically require high applied electric fields (as high as 1.28×10^8 V/m)[2,3] to actuate, which severely restricts their applications.

Another kind of actuator that can perform bending actuation is the thermal bimorph actuator, consisting of two materials with different coefficients of thermal expansion (CTE). The thermally induced actuation mechanism is due to a major mismatch in film properties and CTE of the materials. This kind of actuator has been commonly used in microelectromechanical systems (MEMS), such as thermally excited silicon microactuators,[12] three-dimensional (3-D) MEMS variable optical attenuator,[13] and microactuator for precise track-positioning of optical disk drives.[14] In the past few years, there have been several thermal actuators based on CNTs. For instance, nanoscale trimorph[15] or bimorph[16] devices were developed for thermal actuation by depositing thin metal-oxide or metal films on sidewalls of a multiwalled CNT, and a microcantilever actuator was made from a vertically aligned CNT

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array and polymer composite.[17] Nevertheless, macroscopical scale CNT-based actuators are still rare, and better actuation performance should be sought after for better and more efficient practical application.

In previous studies, we reported an electrothermally actuated CNT/polymer composite.[18] To overcome the drawback of the high-voltage requirement, we incorporated randomly oriented multiwalled CNTs into the polymer to form a highly conductive composite. The actuation mechanism is due to an electrothermal expansion, which differs significantly from that of conventional dielectric elastomer materials. The longitudinal actuator was placed between two copper electrodes. When a low applied voltage of only 1.5 kV/m was applied, thermal expansion and the confinement of the electrodes induce buckling and deflection of the beam upward. Recently, several other CNT-based electrothermal polymer composite actuators were also reported.[19,20]

Lately, we found that embedding super-aligned carbon nanotube sheets (SACNS) into a polymer matrix can remarkably reduce the CTE of the polymer matrix by two orders of magnitude (from 3.1×10^{-4} /K down to 6×10^{-6} /K). Up to now, there are few reports on actuator systems making full use of this unique phenomenon. In this work, based on the discovery, we fabricate an electrothermal bending actuator through a two-step method. The thermally induced actuation is due to a major mismatch in film properties and CTE of the materials. The SACNS/polymer composite actuator is easily operable and can generate exceptionally large bending actuation with controllable motion at very low driving DC voltages (<700 V/m). Furthermore, the actuator can be operated without electrolytes in the air. The striking significance of finding such a high-performance, low-voltage actuator lies in its various potential applications in many fields. A video record demonstrates the practical application of this new actuator (see Supporting Information).

RESULTS AND DISCUSSION

As shown in Figure 1a, the employed CNTs were multiwalled nanotubes arranged almost in parallel to one another along the same direction. As a transparent, flexible and insulating polymer, polydimethylsiloxane (PDMS) was employed as the matrix material. The SACNS/PDMS composites were fabricated through a two-step method: Stacking 50 layers of SACNS on a stainless steel frame and directly placing them on the surface of PDMS before curing. Detailed processes of the fabrication are given in the Methods Section. Compared to our previous work, the method here is much simpler. It does not require intensive ultrasonic or dilute solvent to aid the dispersion of CNTs into the polymer matrix, which simplifies lots of procedures and will highly facilitate large-scale industrial productions.

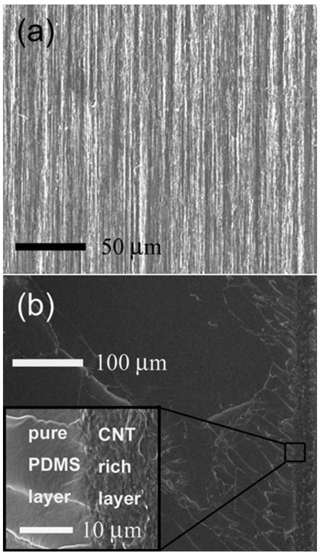

Figure 1. **(a) SEM image showing the surface of a continuous and SACNS. (b) Cross-sectional SEM image of a SACNS/PDMS composite. The right-hand-side layer is the CNT-rich layer, and the left is the pure PDMS layer. Inset: enlarged image of a selected area of the interface.**

The internal structure of the SACNS/PDMS composite is shown in the cross-sectional scanning electron microscopy (SEM) image (Figure 1b). Because SACNS were loosely stacked in order to achieve more contact areas between the CNTs and the polymer matrix, a CNT-rich layer was formed on the surface of the composite, and the sheets were all localized in this region. This CNT-rich layer had a thickness of only 20 μm. Another layer consisting of only pure PDMS was beside the CNT-rich layer, with a much thicker thickness of around 0.75 mm. Two layers were tightly coupled to each other due to our fabrication method shown clearly by the enlarged inset SEM image. In general, the CTEs of two materials in conventional thermal bimorph actuators are at the same order of magnitude ($\sim 10^{-6}$ /K).[12] However, the CTE mismatch between the CNT-rich layer in the direction parallel to CNT alignment and the pure PDMS layer is enormous. The CTE of the CNT-rich layer in the direction parallel to CNT alignment was measured to be 6×10^{-6} /K, which is at the same order of that of multiwalled CNTs (less than 3×10^{-6} /K).[21] This value is two orders of magnitude lower than that of pure PDMS (3.1×10^{-4} /K).[22] Therefore, the actuator structure employing SACNS to form a CNT-rich layer is predictable to perform excellent bending actuation. The mechanism for the dramatic reduction of CTE of PDMS by blending it with super-aligned CNTs is as follows: CNTs are super-aligned in one direction and join end to end through strong van der Waals forces. Therefore, the CTE of super-aligned CNTs in the direction parallel to CNT alignment will be similar to that of multiwalled CNTs (less than 3×10^{-6} /K).[21] When PDMS was blending with super-aligned CNTs, the large specific surface area of CNTs results in large

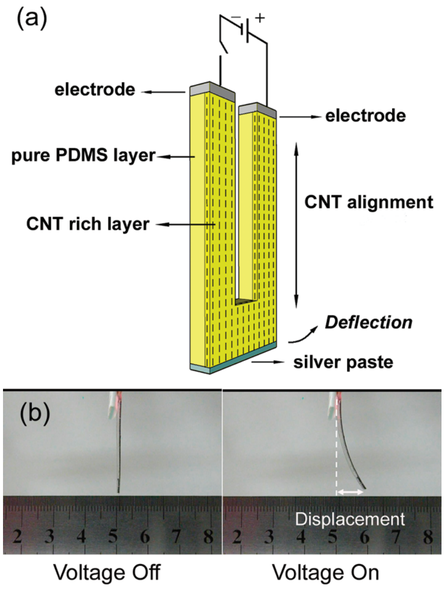

Figure 2. (a) Schematic structure of the SACNS/PDMS composite actuator. The dash lines represent the direction of CNT alignment. (b) Photographs of an actuator without (left) and with (right) an applied DC voltage of 40 V. The unit of the ruler is 0.5 mm.

interfaces between CNTs and the PDMS matrix. Because the CTE of the composite is determined by the thermal expansion of the matrix and the restriction of the reinforcement through interfaces,[23] large interfaces between CNTs and PDMS restrict the thermal expansion of PDMS, which dramatically reduces the CTE of the PDMS in the direction parallel to CNT alignment.

As Figure 2a displays, a U-shape actuator was fabricated in order to study the bending actuation of the SACNS/PDMS composite. The geometry of the U-shape was also reported by Mather[24] and Leng[25] *et al.* in shape memory nanocomposites. The purpose of this design is to fix one end of the actuator (named "fix-end") to two electrodes. Then when the actuator performs a bending actuation, the other end of the actuator (named "free-end") can move freely without the confinement of electrodes. The bending actuation performance of the SACNS/PDMS composite was demonstrated with a suspended geometry as shown in Figure 2b. The actuator was attached to two copper electrodes. A digital camera was used to characterize the bending movement of the actuator, together with an infrared thermometer to monitor the temperature variation.

The current−voltage ($I-V$) curve of the SACNS/PDMS composites actuator is shown in Figure 3a. The linear line indicates ohmic behavior of the composite. Calculation results showed that the CNT loading in the CNT-rich layer of the composite shown in Figure 1b was about 5 wt % and that the overall loading in the entire composite was about 0.1 wt % (see Supporting Information for details). Since the pure PDMS layer is insulated, the electrical conductivity of the CNT-rich layer is calculated to be up to 2280 S/m, due to the continuous and super-aligned structure of the CNT conductive network. And the overall electrical conductivity of the composite was 60 S/m. The former conductivity is two orders of magnitude higher than the reported conductivity of composite prepared by randomly oriented CNT/PDMS composites with 5 wt % CNT loading.[18]

When a DC voltage of 40 V was applied, the current was 78.5 mA, and the actuator began to bend immediately. The voltage was cut off after being applied for 5 s, and the free-end displacement of the actuator reached up to 9.5 mm (Figure 2b). To investigate the actuation mechanism, the displacement and temperature were measured synchronously during the actuation (Figure 3b). It shows that the temperature variation is nearly in line with the actuated displacements. It is known that CNTs are perfect black bodies with high thermal conductivity, exhibiting excellent thermal absorption properties.[26,27] In the actuator, electrical energies absorbed by the CNTs are converted to thermal energy and heat up the entire actuator immediately. Since CTE mismatch between the CNT-rich layer in the direction parallel to CNT alignment and the pure PDMS layer is enormous, the same temperature rise will lead to a phenomenon that the expansion of pure PDMS layer is much larger than that of the CNT-rich layer, which results in an exceptional bending actuation of the structure. The temperature dependence of the displacement is also shown in Figure 3c. It can be seen that the displacement increased rapidly with temperature, which further illuminates that the actuation is related to a thermal effect. Once the voltage was cut off, the free-end returned to its initial position in 60 s.

The maximal displacement was 9.5 mm at the temperature of 98 °C. For the temperature change from 26 °C (room temperature) to 98 °C, the output displacement normalized to the beam length as a function of temperature was 0.44 μm /(100 μm × °C), which is much greater than that of existing bimorph thermal actuators, *e.g.*, 0.15 μm /(100 μm × °C),[17] 0.1 μm /(100 μm × °C),[13] and 0.01 μm /(100 μm × °C).[14]

The electric power density dependence of the displacements was measured and shown in Figure 3d. All data were obtained by inputting a certain electric power for 5 s. It can be seen that by increasing electric power density, more electric energies were input, and then more thermal energies were converted to heat by CNTs rapidly, which resulted in larger displacements. Therefore, the actuator is quite controllable and could be easily controlled through input energy. The actuator is quite reversible as well. The DC voltage of 40 V was applied for 100 cycles (60 s for one cycle), and the actuations were repeatable with nearly the same displacement amplitude, as shown in Figure S2, Supporting Information. A cross-sectional SEM of the actuator

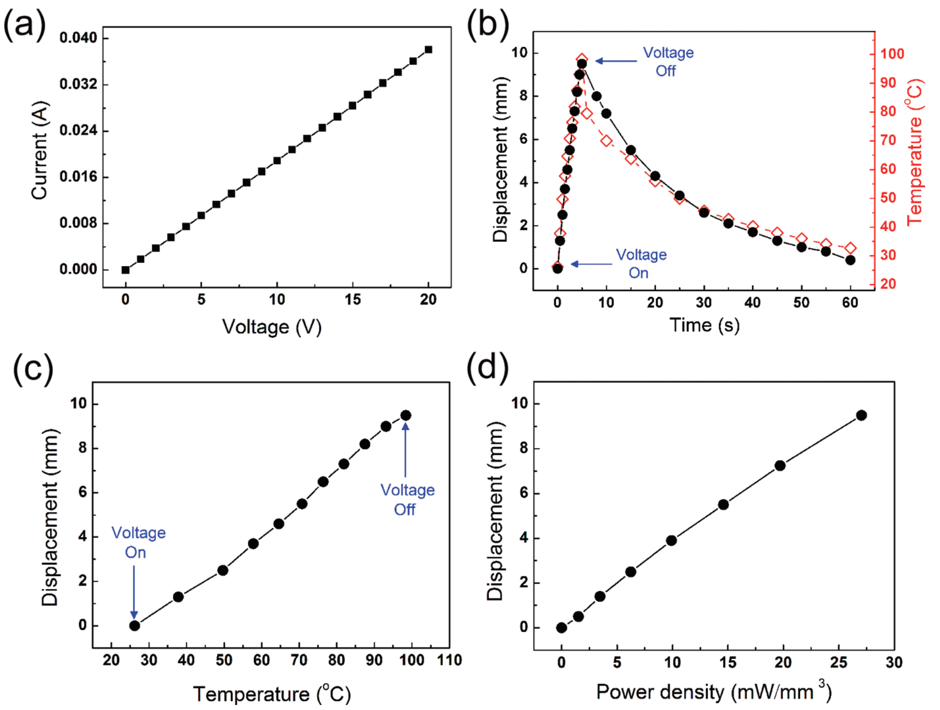

Figure 3. (a) Current−voltage (*I*−*V*) curve of the SACNS/PDMS composite actuator. (b) Displacements (black solid cycle) and temperature (red open diamond) variation of SACNS/PDMS composite actuator with an applied DC voltage of 40 V and current of 78.5 mA. (c) Temperature dependence of the displacements of SACNS/PDMS composite actuator. (d) Power density dependence of the displacements of SACNS/PDMS composite actuator.

after cycling shows that there was no delamination of CNTs from the PDMS matrix (Figure S3, Supporting Information).

Traditional thermal bimorph actuator, consisting of two materials with different CTEs, can perform bending actuation as well. However, the sandwich structure design usually requires additional adhesives and heating layers. The incorporation of these layers often brings in an extra fabrication process and causes delamination which can deteriorate the actuation performance and reduce the working life of the devices.

The reasons and advantages for choosing SACNS/PDMS material system for electrothermal actuation are as follows: (1) As is shown in Figure 1b, even if a visible interface can be observed between the CNT-rich layer and the pure PDMS layer, both layers were tightly coupled to each other due to our fabrication method. Hence, in contrast to traditional thermal bimorph actuators, the present actuator based on SACNS/PDMS material system does not require adhesive layers. Moreover, the high conductivity of SACNS makes the CNT-rich layer as a conductive and heating layer. The unity avoids the delamination of the actuator structure (Figure 1b and Figure S3, Supporting Information). (2) The CTE mismatch between the CNT-rich layer in the direction parallel to CNT alignment and the pure PDMS layer is enormous. We also made another kind of CNT/PDMS composite for comparison, in which the CNTs were not super aligned but randomly oriented. The CTE of this composite was measured to be 3.0×10^{-4} /K,[18] which is nearly the same as pure PDMS.[22] Hence, we can assume that only the incorporation of SACNS can result in enormous mismatches in CTE between two layers. (3) The extraordinary flexibility and mechanical resilience of CNTs and PDMS ensure a long work life of the actuator under repeated bending,[28,29] which is also shown in Figures S2 and S3, Supporting Information.

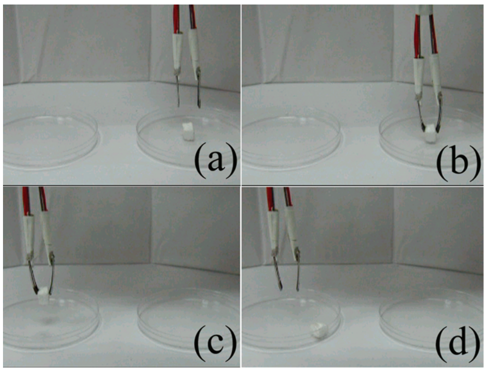

Figure 4. A demonstration of a gripper manipulating a small object: (a) without, (b) with, (c) with, (d) without an applied DC voltage of 35 V.

We fabricated a tiny gripper to demonstrate the potential application of this kind of actuator. Two actuators attached to two sides of the gripper had the same structure as discussed earlier, shown in Figure 2a, but the lengths were both 25 mm. This device was used to manipulate a small object (a piece of plastic foam) with a dimension of around 10 × 10 × 6 mm from one

Petri dish to another (Figure 4). In the beginning, the gripper was open without electrical voltage applied. When a DC voltage of 35 V was applied, the actuators began to bend rapidly, and the free-ends of both actuators were getting closer to each other and finally clamp the small object. Afterward the gripper was moved steadily and placed upon another Petri dish. Then the voltage was cut off, and the gripper was open again, releasing the small object. A video record (see Supporting Information) clearly shows the whole process of the manipulation. The demonstration is quite simple. More improvements will be achieved in our future work. For instance, replacing the PDMS with other polymers to see whether better bending actuation performance could be achieved. By further design and optimization, we believe that this kind of actuator has huge potential in lots of areas.

CONCLUSION

In summary, we have proposed a new kind of bending actuator based on super-aligned CNTs and a PDMS composite. The mechanism of actuation is attributed to an electrothermal effect. The actuator is easily operable and can perform remarkable bending actuation than the existing thermal actuators. It can work in the air at very low-driving DC voltages. This study also demonstrates the potential ability of using this kind of actuator in various applicable areas, such as artificial muscles, microrobotics, microsensors, microtransducers, micromanipulation, microcantilever for medical applications, and so on.

METHODS

Continuous and Super-Aligned Multiwalled CNT Sheets Preparation. Super-aligned multiwalled CNTs arrays were synthesized on a 4 in. silicon wafer in a low-pressure chemical vapor deposition (LPCVD) system by using Fe film as the catalyst and acetylene as the precursor.[30,31] The height of the CNT arrays was around 300 μm. The CNTs were multiwalled nanotubes consisting of about 5−10 concentric nanotube shells having an outer diameter of about 6−20 nm. Continuous and aligned CNTs sheets were directly drawn from these super-aligned CNT arrays by a solid state process.[30−32] The super-aligned CNTs have very clean surfaces, and they are self-assembled by strong van der Waals forces. When pulling the CNTs from the super-aligned array, the force makes the CNTs join end to end, forming continuous and aligned CNTs sheets.

SACNS/PDMS Composite Preparation. The matrix material polydimethylsiloxane (PDMS) rubber (Beijing Hangtongzhou Technology Co., Ltd., China) is a transparent and flexible polymer. It is an electrical insulator (resistivity $>10^8$ $\Omega\cdot$m). Pure PDMS polymer was mixed with tetraethyl orthosilicate cross-link reagent at a weight ratio of 100:6. The SACNS/PDMS composite was fabricated by the following two-step method: (1) Stacking 50 layers of super-aligned and ultrathin continuous CNT sheets on a stainless steel frame. (2) The SACNS together with the frame was directly placed on the surface of PDMS before curing. After curing in an oven at 80 °C for 24 h, solid-state composites were achieved. Afterward the frame was removed from the SACNS/PDMS composite.

Fabrication and Temperature Measurement of the U-shape Actuator. The dimensions of the entire U-shape actuator was 30 × 6 × 0.77 mm (length × width × thickness), while the width of each beam was around 2.5 mm. It was made from strip-shaped SACNS/PDMS composite by cutting off the middle part of it. The free-end of the U-shape actuator was coated with silver paste to enhance conductivity in the direction perpendicular to the CNT alignment. The temperature of the U-shape actuator was measured by a laser sight infrared thermometer (Optris LS) with temperature resolution of 0.1 °C. The temperature data were obtained from the surface of CNT-rich layer. We also measured the temperature of the other surface (pure PDMS layer), and they were nearly the same.

Fabrication of an Ultrathin SACNS/PDMS Composite Film. Because the properties of CNT-rich layer are required, we fabricated an ultrathin SACNS/PDMS composite film to represent the CNT-rich layer and investigated the properties. The ultrathin SACNS/PDMS composite film was fabricated by the following method: (1) Stacking 50 layers of super-aligned and ultrathin continuous CNT sheets on a stainless steel frame. (2) Pure PDMS polymer was mixed with tetraethyl orthosilicate cross-link reagent and ethyl acetate at a weight ratio of 100:6:200. The function of ethyl acetate is to dilute the pure PDMS polymer and avoid the forming of pure PDMS layer. (3) About 10 drops of the liquid mixture were dripped on the SACNS by using a dropper. The liquid mixture could easily spread and wet the entire SACNS, and then the composite film was hung up for 2 h. (4) After curing in an oven at 80 °C for 24 h, solid-state ultrathin SACNS/PDMS composites were achieved. The ethyl acetate was totally volatilized in this process. Afterward the frame was removed from the SACNS/PDMS composite.

The cross-sectional SEM image of the ultrathin SACNS/PDMS composites film (Figure S1, Supporting Information) shows that there were no pure PDMS layers. The SACNS were not concentrated on the surface of the film but uniformly distributed throughout the film. The structure of this ultrathin composite film is the same as the CNT-rich layer in the actuator.

The CTE of CNT-Rich Layer. We measured the CTE of the ultrathin SACNS/PDMS composites film to investigate the CTE of CNT-rich layer.

The dimensions of the ultrathin composite film sample were 68 × 5 × 0.02 mm (length × width × thickness). The length is along the direction in parallel to CNT alignment. The sample was put on a hot plate (CHEMAT KW-4AH-350) with temperature resolution of 1 °C. The formula of CTE was given by

$$\Delta L/L = \alpha\cdot\Delta T$$

where L_0 is the initial length of the sample, ΔL is the increased length, α is the CTE, and ΔT is the increased temperature. Hence, by measuring the length of the sample under different temperatures, we got a series of data of $\Delta L/L$ and ΔT. The slope of $\Delta L/L$ against ΔT curve over a temperature range can also be derived from the data, which is α. The length change was measured by an optical laser distance sensor (Leuze ODSL8) with resolution of 0.01 mm. The temperature was measured by a laser sight infrared thermometer (Optris LS) with temperature resolution of 0.1 °C. The CTE of this ultrathin film in the direction parallel to CNT alignment was measured to be 6×10^{-6} /K, which is at the same scale of that of the multiwalled CNT (less than 3×10^{-6} /K).[21]

We also measured the CTE of the ultrathin SACNS/PDMS composite film in the direction perpendicular to the CNT alignment, which could represent the property of the CNT-rich layer and the CTE of the super-aligned CNT in this direction. The CTE value is 3×10^{-4} /K, which is almost the same as pure PDMS. The reason is that the interaction between CNTs is weak and that the property of PDMS matrix plays a leading role in this direction. Therefore, the CTE of CNT-rich layer and super-aligned CNT in the direction perpendicular to the CNT alignment is strongly different from that in the direction parallel to the CNT alignment, which shows the anisotropic property of the material.

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Supporting Information Available: Additional information for the weight fraction of CNTs in the composite; cross-sectional SEM image of an ultrathin SACNS/PDMS composites film; repeatability of the SACNS/PDMS composite actuator; thermostability of the SACNS/PDMS composite actuator. This material is available free of charge *via* the Internet at http://pubs.acs.org.

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