

## Spongy graphene based bimorph actuator with ultra-large displacement towards biomimetic application

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### Abstract

Bimorph actuators, consisting of two layers with asymmetric expansion and generating bending displacement, have been widely researched. Their actuation performances greatly rely on the differences of coefficient of thermal expansion (CTE) between the two material layers. Here, by introducing a spongy graphene (sG) paper with large negative CTE as well as highly electrical-to-thermal property, an electromechanical sG/PDMS bimorph actuator is designed and fabricated, showing ultra-large bending displacement output under the low voltage stimulation (curvature of about  $1.2\text{cm}^{-1}$  under 10V voltage for 3s), high displacement-to-length ratio ( $\sim 0.79$ ), and vibration motion under AC voltage (up to 10Hz), which is much larger and faster than that of the other electromechanical bimorph actuators. Based on the sG/PDMS bimorph serving as the “finger”, a mechanical gripper is constructed to realize the fast manipulation of the objects under 0.1 Hz square wave voltage stimulation (0~8V). The designed bimorph actuator coupled with ultra-large bending displacement, low driven voltage, and ease fabrication, may open up substantial possibilities for the utilization of the electromechanical actuator in practical biomimetic device applications.

### Full Text

#### Preamble

Spongy Graphene-Based Bimorph Actuator with Ultra-Large Displacement for Biomimetic Applications

Bimorph actuators, consisting of two layers with asymmetric expansion that generate bending displacement, have been widely researched. Their actuation performance greatly relies on the difference in coefficient of thermal expansion (CTE) between the two material layers. Here, by introducing a spongy graphene

(sG) paper with large negative CTE and excellent electrothermal properties, we design and fabricate an electromechanical sG/PDMS bimorph actuator that demonstrates ultra-large bending displacement output under low voltage stimulation (curvature of about  $1.2 \text{ cm}^{-1}$  under 10 V for 3 s), a high displacement-to-length ratio ( $\sim 0.79$ ), and vibration motion under AC voltage (up to 10 Hz), which is much larger and faster than other electromechanical bimorph actuators. Using the sG/PDMS bimorph as a “finger,” we construct a mechanical gripper that can rapidly manipulate objects under 0.1 Hz square wave voltage stimulation (0–8 V). The designed bimorph actuator, featuring ultra-large bending displacement, low driving voltage, and ease of fabrication, may open substantial possibilities for practical electromechanical actuators in biomimetic device applications.

## Introduction

Actuators that convert electrical energy into mechanical energy have received great attention due to their fascinating properties and diverse applications in artificial muscles, switches, microrobotics, manipulators, sensors, and other devices. As an important branch, bimorph actuators have been intensively developed over the past few years because of their simple structure and prominent actuation performance. Their typical hybrid structures consist of two layers with different thermal expansion behaviors, which generate bending displacement under external stimulation due to asymmetric thermal expansion. Therefore, actuation performance is greatly affected by the mismatch in coefficient of thermal expansion (CTE) between the two material layers. Traditional bimorph materials include metals, metal oxides, and silicon, most of which have positive CTE (expanding upon heating). However, they are commonly used in micro or nanoscale actuators, and the generated displacements are restricted by their relatively low CTE difference.

Alternative materials such as carbon nanotubes (CNT) and graphene have been developed recently for their outstanding properties and high actuation performance. A microcantilever bimorph actuator made from CNT and parylene has been reported to operate at a low voltage of 50 V. Subsequently, a macroscopic 30 mm-length bimorph actuator composed of CNT and PDMS was reported to achieve a large displacement of 9.5 mm under 30 V in 5 seconds. Considering that pure CNT has a very small positive CTE, it is possible to enhance displacement performance by replacing CNT with negative CTE materials. Graphene and its derivatives have been studied as new bimorph materials due to their distinctive negative CTE property, which contrasts with conventional materials. However, the intrinsic negative CTE of graphene sheet due to negative in-plane lattice deformation is relatively small. Graphene oxide (GO) paper exhibits a large negative CTE ( $\sim -10 \text{ }^\circ\text{C}$ ) resulting from fast transport of water molecules attached to hydrophilic regions with oxygenated functional groups on GO sheets. Unfortunately, the poor electrical property of GO paper restricts its application, especially for electric actuation, and the reduction of GO to increase its conduc-

tivity would remove the oxygen-containing functional groups and thus diminish the negative thermal expansion.

Therefore, the exploitation of electromechanical bimorph actuator materials with vast CTE difference for large displacement output is highly desirable for the development of electric actuators.

Very recently, we reported a spongy graphene (sG) paper with an internal foldable corrugated structure. Compared with other graphene materials, the sG paper shows low-voltage-induced macroscopic length contraction and large negative CTE ( $\sim -10$  / $^{\circ}\text{C}$ ), mainly due to corrugated structure deformation from interlayer gas expansion. This likely makes the sG paper a potential candidate for designing high-performance electromechanical bimorph actuators. In this paper, by combining the sG paper with PDMS material, which has high positive CTE, we introduce an electromechanical bimorph actuator that exhibits low-voltage-induced ultra-large bending displacement due to the vast CTE difference between the two materials. When applying 10 V DC voltage to this sG/PDMS bimorph, a bending displacement of 15 mm with curvature of about  $1.2\text{ cm}^{-1}$  and strain of 0.41% is achieved in 3 s, which is much larger and faster than other similar reported bimorph actuators. After 10 V is applied for 5 s, the sG/PDMS bimorph further bends with a bending angle of about  $133^{\circ}$ . When applying AC voltage, the bimorph shows reversible vibration motion with frequency up to 10 Hz. An extremely high displacement-to-length ratio is realized. The designed electromechanical bimorph actuator with excellent performance may provide a promising foundation for constructing highly biomimetic devices with multifunctional applications. As a demonstration, mechanical grippers are designed using the sG/PDMS bimorphs as “fingers” to realize fast manipulation of objects (2 s) under low voltage stimulation, and a prototype voltmeter with the sG/PDMS bimorph as the pointer is fabricated to indicate DC voltages ranging from 0 V to 10 V.

## Results and Discussion

The sG paper is fabricated from reduced graphene oxide (RGO) paper by an electrothermal reduction method, in which thermal reduction of oxygen-containing functional groups inside the RGO paper results in rapid gaseous species expansion and thus the formation of internal spongy structures. More detailed fabrication process of the sG paper can be found in our previous work. Figure 1a [Figure 1: see original paper] shows the cross-sectional SEM image of the sG paper. Corrugated structures with fusiform cavities are clearly observed. Because of deformation of the corrugated structures induced by thermal expansion of gas inside the fusiform cavities, the sG paper exhibits extraordinary thermal contraction in the length direction with a large negative CTE of  $-10$  / $^{\circ}\text{C}$ . Moreover, the sG paper has excellent electrical properties. Figure 1b gives the current-voltage (I-V) property of the sG paper. Because of the high electrical conductivity ( $\sim 40$  S/cm) as well as two-dimensional structure, the sG paper can also serve as a heater to quickly convert electric current into thermal

energy, providing thermal stimulation for actuation of thermal response polymer materials. Compared with other materials such as one-dimensional CNT, the two-dimensional graphene has large heating area and can offer more sufficient and efficient heating for the contact interface. Therefore, based on sG paper with these features, an electromechanical bimorph actuator with large bending displacement can be constructed by combining the sG paper layer with a positive-CTE polymer material layer, as shown in figure 1c.

Figure 2a [Figure 2: see original paper] displays the schematic setup of the designed electromechanical bimorph actuator. Here, a U-shape configuration is employed to form a current circuit for electrothermal stimulation, and PDMS is chosen as the polymer layer for its high positive CTE ( $\sim 3 \times 10^{-4} / ^\circ\text{C}$ ) as well as thermal stability. The U-shape sG/PDMS bimorph is connected to voltage such that the two ends of the sG side adhere to the two electrodes with silver conductive adhesive. Due to imbalanced stress in the different layers, the original bimorph is curved toward the PDMS side. Upon applying electric voltage to the bimorph, the sG paper is directly heated and shrinks along the length direction, while the PDMS layer is heated by diffused heat and shows thermal expansion. The vast thermal expansion difference between sG and PDMS layers is expected to cause ultra-large bending displacement for the bimorph under electric stimulation.

Figures 2b–2d show side-view optical images of the sG/PDMS bimorph under applied 10 V voltage for 0 s, 3 s, and 5 s (a supplementary video record of the cycled actuation of the sG/PDMS bimorph under 10 V for 5 s is also provided in the Supplementary Information). The bimorph shows slight bending toward the PDMS side without voltage (figure 2b). When applying 10 V voltage to this 27 mm-length sG/PDMS bimorph for 3 s, it bends toward the sG side to generate a displacement of 15 mm with large curvature of about  $1.2 \text{ cm}^{-1}$  and strain of 0.41% (figure 2c). Here the displacement is measured by the ruler below, which is captured from the camera record, as shown by the white dotted line in figure 2c. This can be used for measuring displacement that exceeds the range of the laser displacement sensor. When 10 V voltage is applied for 5 s, the bimorph further bends upward with a bending angle of roughly  $133^\circ$ . The generated actuation motion of the sG/PDMS bimorph is much larger and faster than that of reported electrical bimorph actuators under similar temperature. Because the bending displacement of the bimorph is related to its length, displacement-to-length ratio (D/L) is adopted to evaluate the performance of bimorph actuators with different lengths.

Table 1 summarizes the maximum displacement and D/L of various electrical bimorph actuators under electrical voltage stimulation reported in previous literature. Compared with other bimorph actuators, the sG/PDMS bimorph shows a much higher D/L ( $\sim 0.63$ ) under comparable temperature by applying much lower voltage. For the bimorph with shorter length (19 mm), D/L  $\sim 0.79$  has even been achieved under 0.05 Hz 10 V square wave voltage stimulation.

For the purpose of investigating the bimorph actuation process, the real-time

displacement and temperature variation of the sG/PDMS bimorph with cycled voltage on and off are simultaneously monitored, as shown in figure 3a [Figure 3: see original paper]. Because the displacement under 10 V voltage has exceeded the measuring range of the laser displacement sensor and cannot be wholly recorded, the driven voltage for the bimorph actuator is lowered to 5 V to obtain better measurement of the actuation. When applying 5 V voltage (gray shadow) on the sG/PDMS bimorph for 60 s, the generated bending displacement increases to the maximum and holds until the voltage is off (black triangle in figure 3a). After the voltage is off, the displacement recovers to its original status. For the two actuation cycles, the displacement curves repeat very well. We have also tested the bimorph actuator for 4 cycles. The displacement is still quite reversible and repeats well, indicating that the actuator remains elastic. The simultaneously generated temperature variation during the actuation process (red circle in figure 3a) is almost consistent with that of the displacement, further illuminating the electric-induced thermal mechanism. Furthermore, the influence of environmental temperature on actuation performance is also examined, as shown in figure S1. For environmental temperatures from 22 °C to 25 °C, the actuated displacements under 4 V voltage are almost the same, revealing that small environmental temperature changes may not greatly affect the electromechanical actuation of the bimorph.

As the actuation is determined by the applied electric power-generated Joule heating, the maximum displacement as a function of input electric power is measured and displayed in figure 3b. With increasing electric power, more thermal energy is converted, resulting in larger displacement. The generated temperature versus input electric power and maximum displacement versus temperature are also provided in figure S2. As bimorph actuation depends on thermal contraction of the sG layer as well as thermal expansion of the PDMS layer, the PDMS layer is a key factor for actuation performance, and the thickness of the PDMS layer can greatly affect bimorph actuation behavior.

Figure 3c shows the displacement of the sG/PDMS bimorph under 5 V input voltage with different PDMS thicknesses from 80  $\mu\text{m}$  to 450  $\mu\text{m}$ . It can be seen that with decreasing PDMS layer thickness, displacements increase faster before reaching the maximum. For the bimorph with thinner PDMS thickness, a larger maximum displacement can be generated. However, the maximum displacement for the 80  $\mu\text{m}$  PDMS layer is much smaller than that for the 130  $\mu\text{m}$  PDMS layer. Here we propose an explanation. Decreasing the thickness of the PDMS layer has several effects on tip displacement: (i) it results in better heat transfer and a lower temperature gradient across the PDMS layer. Since the temperature gradient would cause the actuator to bend away from the sG paper and counteract the actuator performance, a lower temperature gradient in a thinner PDMS layer enhances displacement; (ii) the thinner the PDMS layer, the easier it is to be bent by the sG paper layer; (iii) however, a PDMS layer that is too thin does not have sufficient strength to bend the sG paper layer and would follow the contraction of the upper layer without being able to bend the structure. Due to these opposite effects (i and ii vs. iii),

an optimum value for PDMS thickness could be found to achieve maximum displacement. Moreover, an equation for displacement as a function of PDMS thickness is also deduced from a simple bimorph cantilever model (figure S3) to further confirm this explanation. Detailed assumptions and derivation process are provided in the Supplementary Information. From the equation we find that displacement is not a monotonic function of thickness. There should be a moderate PDMS thickness for high-performance bimorphs, neither too thin nor too thick. Considering the analysis and experimental results, the bimorph with PDMS layer thickness of 130  $\mu\text{m}$  is likely an optimal choice and is used unless otherwise mentioned.

Furthermore, to verify the effect of the sG paper on bimorph actuation performance, a comparison of actuated displacement between sG/PDMS and RGO/PDMS bimorphs under the same 5 V voltage is presented (figure 3d). In contrast to the RGO/PDMS bimorph (black curve), the bending displacement for the sG/PDMS bimorph (red curve) is much larger, revealing the significant effect of the sG paper with high negative CTE on bimorph actuation performance.

When applying AC voltage, the bimorph can be driven to generate reversible vibration motion. Because it is difficult to record the bending displacement of the 27 mm-length sG/PDMS bimorph under 10 V stimulation using the laser displacement sensor, the bimorph length is shortened to 19 mm. The vibration displacement of the 19 mm-length bimorph actuator under 5 Hz square wave voltage (0–10 V) is shown in figure 4a [Figure 4: see original paper]. It exhibits 5 Hz vibration with displacement amplitude of 0.5 mm, leading to a D/L of 0.026. Although slight displacement fluctuation exists, which may be due to plastic deformation in the actuator, the vibration is quite reversible with nearly the same amplitude for more than 200 cycles, as shown in figure S4. Figure 4b shows the vibration amplitude of the 19 mm-length bimorph actuator as a function of frequency under square wave voltage (0–10 V). Under 0.05 Hz 10 V stimulation, the bimorph actuator generates displacement amplitude of 15 mm, leading to a D/L of 0.79 and strain of 0.6%. With increasing frequency, the vibration amplitude gradually reduces. Vibration with frequency up to 10 Hz can still be observed; however, frequencies exceeding 10 Hz are not detected, revealing the high-frequency vibration limitation for the sG/PDMS bimorph.

According to the thermal actuation mechanism, the maximal vibration frequency of the actuator is determined by its heat generation and dissipation rate. For macroscale electrically induced thermal actuators, the heating and cooling rates lag behind the rate of current change at frequencies higher than 10 Hz, which leads to disappearance of temperature variation and thus vibration. However, by employing microfabrication technology including photolithography, the dimensions of thermal actuators may be decreased to the microscale with lengths of hundreds of micrometers, thus greatly enhancing the rate of heat dissipation and the frequency of generated actuation due to the increased surface-to-volume ratio. Furthermore, the reliability and stability of the sG/PDMS bimorph is ex-

amed by applying 0.5 Hz square wave voltage (0–5 V) for about 10,000 cycles. No significant performance degradation is observed (figure S5), indicating good stability.

We also study the efficiency of actuation. Because of large heat dissipation, these electrothermal actuators have low energy efficiency compared with other actuator types. Under 10 V stimulation, the efficiency of the sG/PDMS bimorph actuator is estimated to be  $\sim 0.001\%$ , which is on the same order as conventional thermal bimorph actuators. Therefore, much work is still needed to improve the efficiency of electrothermal bimorph actuators.

The sG/PDMS bimorph with ultra-large displacement response to low electric voltage may provide a basis for various device designs and applications. As a demonstration, a “bi-finger” mechanical gripper is fabricated using two sG/PDMS bimorphs as “fingers,” which can quickly grab and release objects with applied voltage on and off. As shown in figures 5a–5f, small objects (plastic foams) with dimensions of about  $10\text{ mm} \times 8\text{ mm} \times 7\text{ mm}$  and weight of 6 mg are placed in a petri dish, and the mechanical gripper manipulates them under applied 0.1 Hz square wave voltage (0–8 V). When driven voltage is applied for 2 s, the “fingers” in the gripper close and the object is tightly grabbed and picked up (figure 5b). After that, the object is quickly moved out of the petri dish and held steadily in the air (figures 5c–5e). When the voltage is off, the “fingers” start to open and put the object down after 2 s, enabling fast object manipulation. A video record of this fast manipulation process by the “bi-finger” mechanical gripper is also provided in the Supplementary Information.

Further, to demonstrate the possibility of constructing more sophisticated biomimetic devices, a “tri-fingered” mechanical gripper with three sG/PDMS bimorphs in rotational symmetry is also fabricated, with the free ends of the bimorphs closing at one point under 0.1 Hz 10 V electric stimulation (figure 6a [Figure 6: see original paper]), and can be employed to grab objects with larger dimensions ( $15\text{ mm} \times 13\text{ mm} \times 12\text{ mm}$ ) and heavier weight (40 mg) under 10 V (figure 6b). Considering the weight of the “bi-finger” gripper is about 36 mg, this gripper can grab objects slightly heavier than the gripper itself.

In addition, for another application example, a simple prototype voltmeter with the sG/PDMS bimorph as the pointer is also fabricated, as shown in figure S6a. After calibrating the pointer displacements under different voltages and labeling the corresponding voltage scale, the voltmeter can be used to sense DC voltages ranging from 0 V to 10 V. The upper part in the images is the electric power supply with the number on its screen displaying the output voltage. Once the voltmeter is connected to the circuit, output voltages of 4 V, 8 V, and 10 V can be indicated by the sG/PDMS pointer, respectively (figures S6b–S6d).

## Conclusions

By employing sG as the large negative CTE material and PDMS as the positive CTE material, we designed an sG/PDMS bimorph electromechanical actuator

that exhibits low-voltage-driven ultra-large bending displacement. By applying 10 V to the bimorph for 3 s, a displacement of 15 mm with curvature of  $1.2 \text{ cm}^{-1}$  and strain of 0.41% is generated. A D/L as high as 0.79 can be realized for the bimorph actuator. Based on sG/PDMS bimorphs that mimic fingers, mechanical grippers are constructed to rapidly grab, move, and release objects. The sG/PDMS bimorph with simple configuration, ease of fabrication, and ultra-large displacement in response to low voltage may provide a basis for various device designs and practical applications in biomimetic robotics, artificial muscles, sensors, and other fields.

## Experimental

### Materials

Natural graphite flake (325 meshes, 99.8%, ABCR GmbH & Co. KG) was purchased from Sigma Aldrich. Phosphorous oxide (purity 98%) and potassium peroxydisulfate (purity 97%) were purchased from Alfa Aesar. HI acid was obtained from Sinopharm Chemical Reagent Co. PDMS material was commercially purchased (Sylgard 184 silicone elastomer, Dow Corning, USA), which consists of A (silicone elastomer liquid base) and B (silicon elastomer curing agent) viscous liquid components. When the base and curing agents are thoroughly mixed in a 10:1 weight ratio, the cross-linking reaction occurs. After that, the viscous PDMS mixture should be placed in vacuum at room temperature to remove air cavities and can be cured as a transparent elastomer at  $80 \text{ }^\circ\text{C}$  for 2 h. All other chemicals were obtained from Sinopharm Chemical Reagent Co., Ltd, and  $18 \text{ M}\Omega$  Milli-Q water was used in all experiments.

### Preparation of RGO/PDMS Bimorph Paper

GO was synthesized from natural graphite flake according to a modified Hummers method. GO paper was fabricated by casting aqueous GO dispersions onto a glass slide and then drying in an oven at  $60 \text{ }^\circ\text{C}$ . RGO paper was fabricated by immersing GO paper into HI acid solution for 10 min. After that, the RGO paper was repeatedly rinsed with ethanol to remove residual HI acid and dried at  $40 \text{ }^\circ\text{C}$ . The RGO/PDMS bimorph paper was fabricated as follows: the RGO paper was placed on a glass substrate, and viscous PDMS mixture was coated onto the RGO paper and then cured in an oven at  $80 \text{ }^\circ\text{C}$ . After that, the dry RGO/PDMS bimorph was peeled off from the substrate to obtain a freestanding bimorph paper. The thickness of PDMS could be tuned by controlling the amount of liquid mixture on the RGO paper. In this work, the bimorph actuator with PDMS layer thickness of  $130 \text{ }\mu\text{m}$  was used unless otherwise mentioned.

### Fabrication of sG/PDMS Bimorph Actuator

The sG/PDMS bimorph actuator was fabricated on the basis of the RGO/PDMS bimorph paper. First, the RGO/PDMS bimorph paper was cut into strips with dimensions of  $27 \text{ mm} \times 7 \text{ mm}$  (length  $\times$  width). The middle part was

cut off from the strips to shape them into a U-shape bimorph. After that, a threshold voltage (10 V) was applied to the RGO side of this bimorph to generate Joule heating, which causes formation of sG from the RGO paper by the electrothermal reduction method. Thus, the U-shape sG/PDMS bimorph actuator was obtained.

### Characterization

For actuation measurement, the bimorph actuators were connected to electrodes at both ends to form an electric circuit. A laser displacement sensor (LK-G80, Keyence) was positioned at the left side of the bimorph actuator to measure bending displacement. For displacements exceeding the range of the laser displacement sensor, a ruler and digital camera could also be used to capture the displacement. Temperature change during the actuation process was measured using an infrared thermometer. GWINSTEK PSM-3004 programmable power supply and CHI600C electrochemical workstation were used for providing DC and AC voltage, respectively. Scanning electron microscopy (SEM) images were obtained with a Hitachi S-4800 field emission scanning electron microscope.

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### Notes and References

† Electronic Supplementary Information (ESI) available: [Video records of the cycled actuation of sG/PDMS bimorph under 10 V for 5 s and the manipulation of objects by the mechanical gripper under 0.1 Hz 8 V voltage, and optical images of a prototype voltmeter based on sG/PDMS bimorph]. See DOI: 10.1039/b000000x/

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