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RESEARCH ARTICLE

Low-Voltage Activating, Fast Responding Electro-thermal Actuator Based on Carbon Nanotube Film/PDMS Composites

Mohamed Amine Aouraghe[1,2] · Zhou Mengjie[1,2] · Yiping Qiu[1,2] · Xu Fujun[1,2]

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Abstract
The electro-thermal actuators (ETA) are smart devices that can convert electric energy into mechanical energy under electro-heating stimulation, showing great potential in the fields of soft robotics, artificial muscle and aerospace component. In this study, to build a low-voltage activating, fast responding ETA, a robust and flexible carbon nanotube film (CNTF) with excellent electrical and thermal conductivity was adopted as the conductive material. Then, an asymmetric bilayer structured ETA was manufactured by coating a thin layer of polydimethylsiloxane (PDMS) with high coefficient of thermal expansion (9.3×10^{-4} °C^{-1}), low young's modulus (2.07 MPa) on a thin CNTF (~ 11 µm). The as-produced CNTF/PDMS composite ETA exhibited a large deformation (bending angle ~ 324°) and high electro heating performance (351 °C) at a low driving voltage of 8 V within ~ 12 s. The actuated movement and the generated heat could be controlled by adjusting the driving voltages and showed almost the same values in 20 cycles. Furthermore, the influences of the PDMS thickness and driving voltage on CNTF/PDMS composite ETA performance were systematically investigated. The CNTF/PDMS soft robotic hand which can lift 5.1 times and crab 1.3 times of its weight demonstrated its potential capability.

Keywords Carbon nanotube film · PDMS · Actuator · Multifunctional composite · Soft robotic

Introduction

Electro-thermal actuators (ETA) are smart devices that can convert electric energy into mechanical energy. Due to their easy fabrication process, low cost and high performance, ETA received increasing attention into several applications such as artificial muscles, micro-pumps, soft robotic and so forth [1–4]. Usually, ETAs are composed at least with two components: a conductor to generate heat by joule heating effect and a polymer to expand after heated by the conductor. Conductive metals such as aluminum foil and copper are widely adopted to make actuators for their high electrical conductivity. Keiichi et al. designed and fabricated a bimorph actuator based on multi walled carbon nanotube (MWCNT), epoxy and aluminum foil [5] that could be activated at a low voltage of 6 V. However, the resulted bending displacement was limited to 10 mm due to the rigid nature of aluminum foil.

To overcome the rigidity and heavy weight of metals, the conductive nano-assemblies with the flexible and low density such as graphene oxide (GO) [6], silver nanowires (AgNws) [7], or carbon nanotube (CNT) [8] are drawn much attention to build the ETA. Some studies showed that their high electrical, thermal conductivity and mechanical properties allow the efficient actuation and robustness to the ETA [9–11]. Zhang et al. developed a fast trigged ETA based on graphene and polyimide (PI) film which can achieve a large bending angle of 270° [12]. However, the bending movement and the limited temperature (130 °C) proposed graphene/PI ETA was reached at a high applied voltage of 65 V with large power consumption, due to the unsatisfied conductivity of the graphene sheet. Yao et al. proposed a bimorph actuator based on silver nanowires (AgNws) that can extremely bend (720 °C) at a really low voltage of 4.5 V [7]. However, the proposed bimorph actuator took a long time to reach

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✉ Xu Fujun
fjxu@dhu.edu.cn

1 Shanghai Key Laboratory of Lightweight Composite, Donghua University, Shanghai 201620, China

2 College of Textiles, Donghua University, Shanghai 201620, China

the steady state bending angle (~ 40 s) and to recover to the initial shape (60 s). Similarly, an ETA based on MWCNT powder and polymer composites (silicone and polyurethane) also spent a relatedly long time ~ 50 s to reach its bending steady state [8]. According to the actuating mechanism, the ideal asymmetric bilayer structured ETA structure should possess the conductive layer with superb electrical and thermal conductivity, flexibility and stiffness as well as the polymer layer with a high coefficient of thermal expansion (CTE), low specific heat capacity and bending modulus. In addition, the good interfacial thermal transmission from the conductive layer to the polymer layer is also a critical issue to consider.

In this study, the thin (~ 11 µm) and robust CNT film (CNTF) manufactured by floating catalyst chemical vapor deposition with excellent electrical conductivity (~ 1000 S/cm) and thermal conductivity [~ 80 W/(m.k)] was adopted as a conductive layer to build a high performance ETA [13, 14]. The polydimethylsiloxane (PDMS) with high CTE (9.3×10^{-4} °C^{-1}), low glass transition temperature (Tg = − 125 °C) and young's modulus (2.07 MPa) was selected as polymer layer (Figure SI 1a and b), which will facilitate the expansion and deformation of PDMS under heat. Therefore, a low driving voltage, lightweight and fast response ETA was designed and manufactured by coating a thin layer of PDMS on the thin CNTF (~ 11 ± 1.5 µm). The electrical thermal behaviors and actuating performance of the CNTF/PDMS composite ETA were tested. The effects of the thicknesses of PDMS layer, driving voltages and workability in extremely low temperatures were systematically investigated. In addition, a soft robotic finger based on the CNTF/PDMS composite ETA triggered by a low electric power was demonstrated.

Experimental

Materials and Fabrication of CNTF/PDMS Composite ETA

CNTF was fabricated by floating catalyst chemical vapour deposition (FCCVD) method as reported previously [13]. PDMS and curing agent (Sylgard 184 Silicone Elastomer Kit, Dow Corning) were mixed in 1/10 ratio and coated on CNTF with a scalable lame blade to control its thickness. The CNTF/PDMS with different thickness was cured at 100 °C for 35 min. The resulted composite was cut into U shape with a specific dimension to achieve a homogeneous circular bending movement around the horizontal axe of the actuator. Copper tape was soldered to CNTF/PDMS composite edges with silver paste to minimize the electrical resistance between the actuator and power source. Figure 1a summarized the actuator fabrication process.

Characterization

The CNTF surface morphology, thickness and its adhesion to PDMS were observed by a Scanning Electron Microscope (SEM, Hitachi TM3000, and 5 kV) and field emission SEM (FESEM, Hitachi S4800, and 5 kV). The chemical structure of CNTF and PDMS were investigated by the Fourier transform infrared (FTIR). The weight loss in the air (O_2) of CNTF and PDMS were investigated by the thermal gravimetric analysis (TGA, Netzsch TG 209F1). Tensile tests of CNTF (10 × 5 mm) were measured on the XQ-2 tensile test machine (Shanghai Xusai Instrument Co, China) with a 3 N load sensor and a crosshead speed of 0.5 mm/min. Power source ODP3031 was utilized to supply different voltage and activate the CNTF/PDMS composite ETA. The resulted bending angle was recorded by a Nikon camera D500. Thermal camera FOTRIC 225 was utilized to measure the temperature generated from the ETA under different voltages.

Results and Discussion

CNTF/PDMS Composite ETA

A U-shaped ETA based on CNTF coated with different PDMS thicknesses was fabricated and studied (Fig. 1a). After connecting with the applied voltages, the CNTF/PDMS composite ETA converts the electrical energy to mechanical bending movement by expanding the PDMS polymer coated on CNTF surface via joule heat effect, as schemed in Fig. 1b. The uniform bending movement was due to the good interfacial adhesion between PDMS and CNTF (Fig. 1c). The surface morphology image (Fig. 1d) revealed that the CNTF is mainly formed by a porous entanglement of CNT fibers which facilitate the propagation of PDMS on the CNTF surface.

Material Characterizations

FTIR reveals that CNTF is mainly formed by CH, CO, CH_2 and OH groups as shown in Fig. 2a. After coating PDMS on CNTF, the FTIR of the composite in Fig. 2b, shows an apparition of silicone groups such as $SiCH_3$, SiO and $Si(CH_3)_2$. The atomic structure of the CNTF and

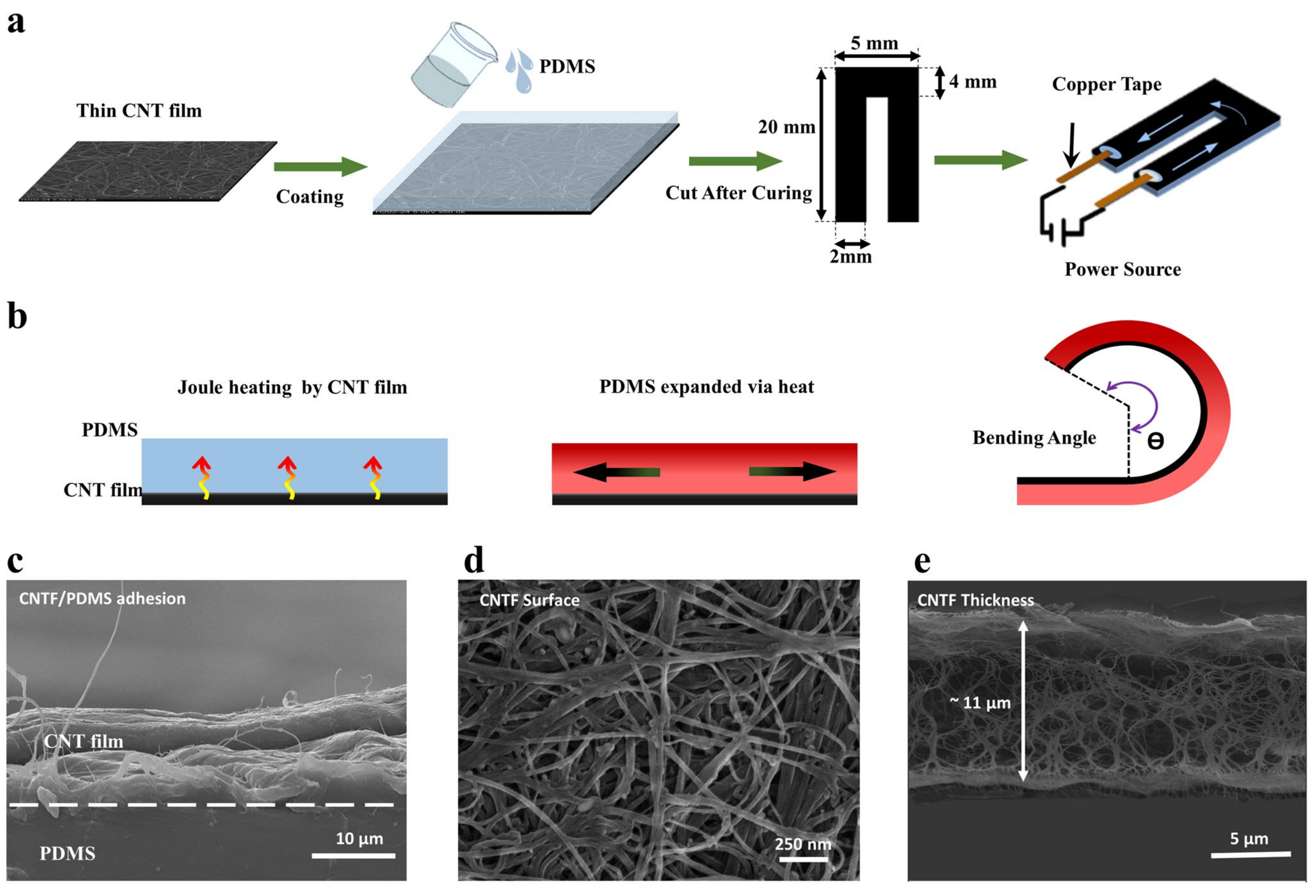

Fig. 1 Schematic of **a** CNTF/PDMS composite ETA fabrication process and **b** its bending process. SEM images of **c** CNTF and PDMS interface adhesion, **d** CNTF surface and **e** cross section

PDMS have a great effect on the CNTF/PDMS thermodynamic properties. The TGA result in Fig. 2c shows that the weight loss of PDMS stands until 300 °C before a total degradation at ~ 380 °C. On the other hand, an important weight loss of ~ 20% of CNTF was observed (Fig. 2c) before a degradation at ~ 480 °C. The important difference in weight loss was due to the amorphous CNTs and remained catalysts present in the film structure, which are oxidized during the TGA process. The TGA results revealed that the CNTF/PDMS composite can stand temperature up to 380 °C.

To further study the thermal stability of CNTF, joule heating behavior of CNTF was further tested. The CNTF of 2 cm × 0.5 cm converts the applied voltage of 2.0 V to ~ 210 °C, within 1 s as shown in Fig. 2d. Furthermore, the temperature remained stable during the applied voltage. After switching off the power source, the CNTF cooled to room temperature within 5 s showing the fast heat dissipation of the CNTF to the surrounding environment. The excellent electrical heating (E-heating) behavior was due the high electrical conductivity of CNTF (~ 1000 S/cm) and the good entanglement and junctions of CNT fibers (Fig. 1d) which leads to a fast trigger and dissipation of heat to the surrounding environment. Moreover, the CNTF exhibits excellent stability and reliability after 12 h ON/OFF cycles, with a frequency of 1 min ON and 30 s off, at an applied voltage of 1 V. The steady state temperature remained stable during the cyclic voltage as shown in Fig. 2e which is essential in CNTF/PDMS composite ETA actuation controllability. In addition to the excellent electro-thermal performances, the proposed CNTF exhibits high mechanical properties with a tensile strength of ~ 130 MPa and an elongation at break of 22% (Fig. 2f), which provides to the CNTF/PDMS composite ETA strength and flexibility. The high tensile strength was due to the packing structure and the preferential alignment of CNT fibers along the production direction, which beards the load during traction and external forces [15].

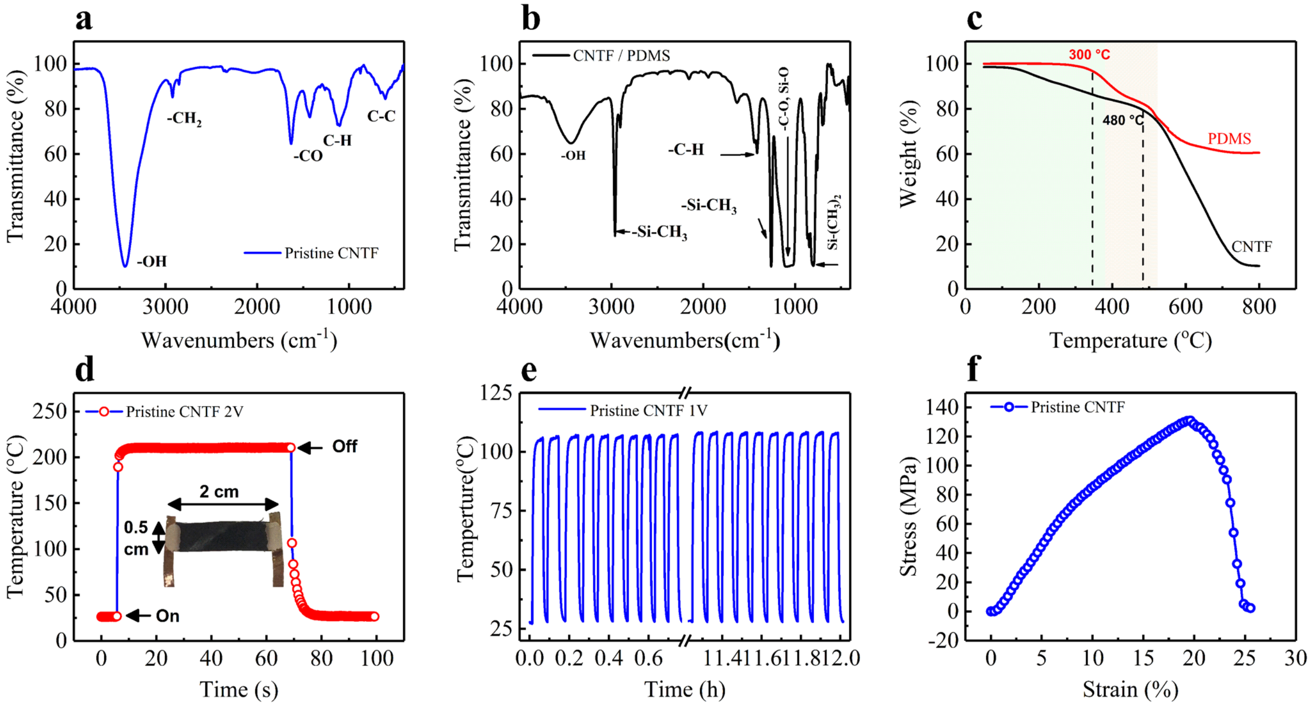

Fig. 2 FTIR results of **a** CNTF and **b** CNTF/PDMS composite. **c** TGA results of CNTF and PDMS. E-heating properties of **d** CNTF at a constant voltage of 2 V for 1 min and **e** cyclic ON/OFF for 12 h under 1 V. **f** Tensile behavior of CNTF

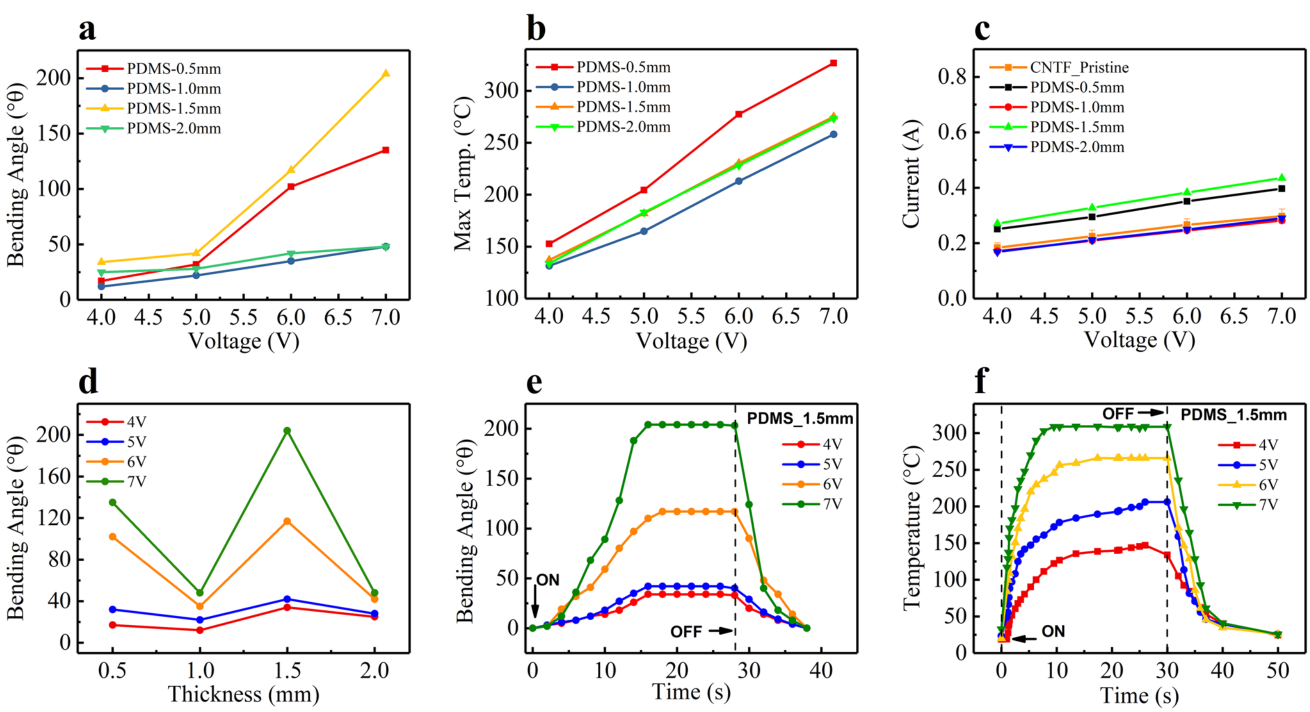

Fig. 3 **a** CNTF/PDMS composite ETA bending responses, **b** resultant temperatures and **c** currents with different applying voltages. **d** CNTF/PDMS composite ETA bending angle versus thickness. **e** Bending and **f** temperature profiles of CNTF/PDMS composite ETA

Actuating Performance of the CNTF/PDMS Composite ETA

The CNTF/PDMS composite ETA bending angle increased systematically with the increment of the applied voltage regardless of the PDMS thickness as shown in Fig. 3a. The bending angle of the actuator with 0.5 mm PDMS in thickness, increases from 17° to 173° at an applied voltage of 4 V and 7 V respectively, due to the temperature increment from 152 °C to 326 °C and electrical conductivity from 0.25 A to 0.4 A as shown in Fig. 3b, c respectively. With the temperature increment, the PDMS polymer tends to expand more due to its high CTE which leads to a large bending angle of the CNTF/PDMS composite ETA. As show in Fig. 3d, the PDMS thicknesses showed a great influence on the CNTF/PDMS composite ETA actuation. Under the same applied voltage of 7 V, the actuator coated with 0.5, 1.0, 1.5, 2.0 mm PDMS in thickness exhibit a bending angle of 135°, 48°, 204° and 48° respectively. Because the thin PDMS layer can be heated up and expand quickly but the expansion force is limited, while he thick PDMS layer can provide large expansion force but difficult to be heated and thus reach low temperature as well as limited bending deformation. There optimum amount of PDMS coating was 1.5 mm which exhibits the large bending angle as demonstrated in Fig. 3d.

The steady-state and bending angle profile of the CNTF/PDMS composite ETA with 1.5 mm in PDMS thickness under various applied voltages were monitored and depicted in Fig. 3e. The bending angle of the CNTF/PDMS reached 34°, 42°, 117° and 204° under the applied voltage of 4, 5, 6 and 7 V respectively, which demonstrate that the low voltage was capable to trigger the mechanical bending movement of the CNTF/PDMS composite ETA. In addition, the bending movement reached its steady state within 15 s and remained stable during the applied voltage (Fig. 3e). On the other hand, and after switching OFF the power source, the CNTF/PDMS composite ETA recovered totally to its initial shape within 10 s. The fast actuation was mainly due to the excellent electro heating performances of the CNTF/PDMS composite and the relatively high electrical power. As depicted in Fig. 3f, the CNTF/PDMS composite ETA convert efficiently the applied voltage of 4, 5, 6 and 7 V to high temperatures of 135, 190, 266 and 308 °C within few seconds.

Stability of the CNTF/PDMS Composite ETA

The actuating performance of the CNTF/PDMS composite ETA with a higher applied voltage of 8 V was observed as shown in Fig. 4a, b. The bending movement was trigged within 4 s and reached its maximum bending movement (324°) after 12 s as observed from the optical images in Fig. 4a. The instant actuation and high bending angle were achieved from the extremely high temperature of CNTF/PDMS composite ETA (351 °C), which approaches the operational limit temperature of PDMS obtained previously from the TGA result (380 °C) indicating the practical limit of the CNTF/PDMS composite ETA. In addition, the temperature generated from the ETA was evenly distributed as observed from the thermal images of the side and top of the CNTF/PDMS composite ETA (Fig. 4b) which engendered a uniform bending movement.

In comparison to the proposed CNTF/PDMS composite ETA, Table 1 summarized the performances of other soft actuator-based nanomaterials such as graphene, carbon nanotube powder and silver nanowires. Most actuators presented in Table 1 based other nanomaterials required high voltage to reach their steady state bending angle and exhibited low E-heating performances. However, the proposed CNTF/PDMS composite ETA exhibit both high thermal response under low voltage and reach its steady state bending angle within few seconds, demonstrating the excellent potential of the CNTF/PDMS composite ETA as a multifunctional soft actuator.

The CNTF/PDMS composite ETA bending movement repeatability, reliability, stability, and controllability were investigated. In Fig. 4c, the CNTF/PDMS composite ETA generated the same bending movement ~ 100° after 20 times cyclic voltage of 6 V On/Off which exhibits the repeatability and stability of the proposed actuator. In addition, bending time and recovery kept stable, 16 s and 7 s, after 20 times cyclic voltage as demonstrated in Fig. 4d. The CNTF/PDMS composite ETA exhibited a stable bending response in time with precise movement, which is crucial in practical application.

CNTF/PDMS Composite ETA Application

To demonstrate the potential applications of the proposed actuator in extremely low temperatures, the fabricated CNTF/PDMS composite ETA was immersed into water and frozen to make a surrounding ice block on it. The applied voltage of 8 V to the CNTF/PDMS composite ETA activated the bending movement and melt the surrounding ice as depicted in Fig. 5a. Due to the high E-thermal conversion of the actuator, the ice on the actuator melt within 20 s and the actuator bent to 110° after 28 s as shown in Fig. 5b. In addition, a soft robotic finger based on the CNTF/PDMS composite ETA that can hold and crab objects was fabricated and tested. The CNTF/PDMS composite ETA can grab 1.3 times and carry 5.1 times its

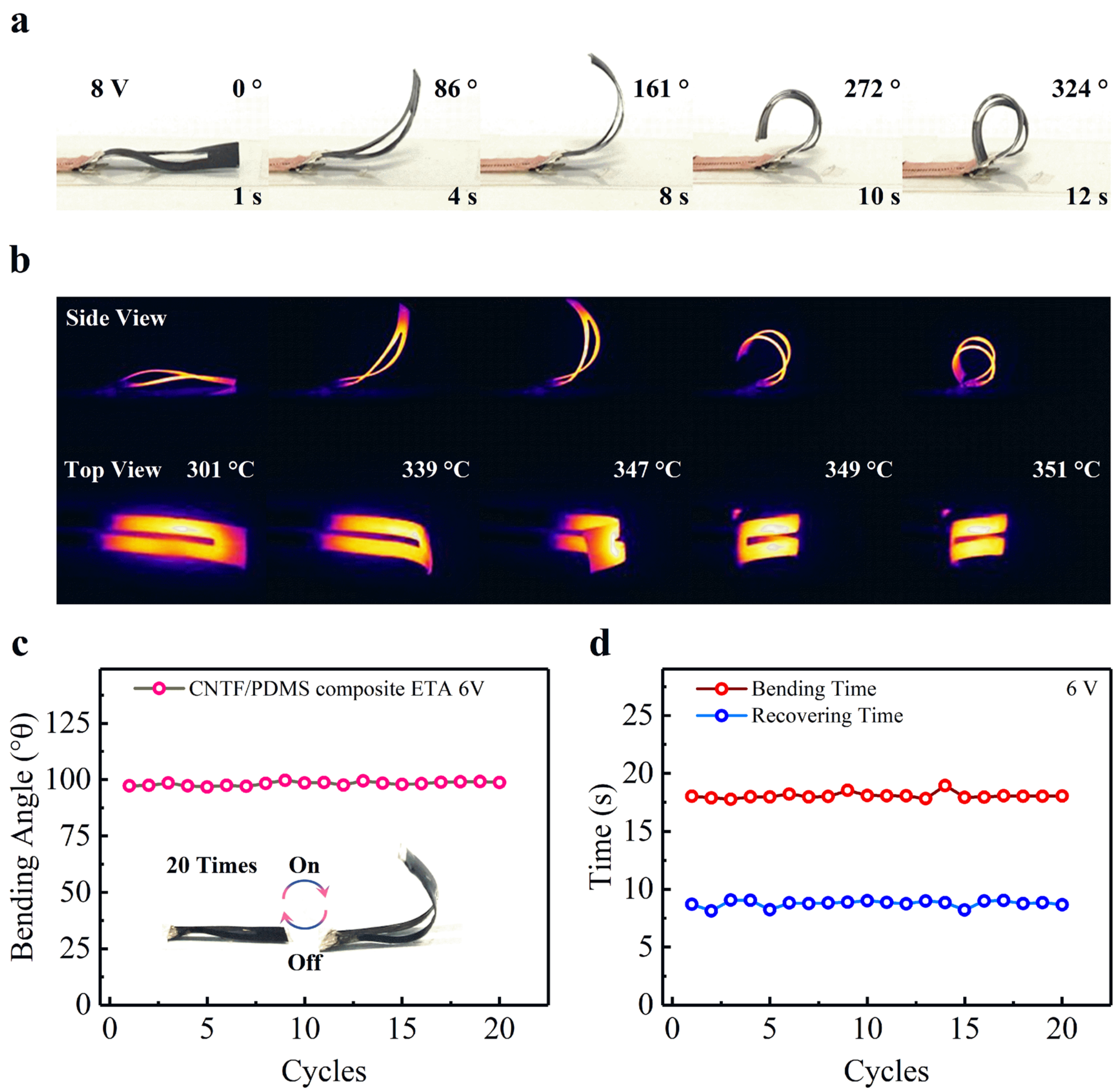

Fig. 4 **a** Optical and **b** thermal images of the CNTF/PDMS composite ETA bending movement under the applied voltage of 8 V. **c** Cyclic bending behavior of the CNTF/PDMS composite ETA after 20 times On/Off under the applied voltage of 6 V and its **c** bending time and recovery time

weight as shown in Fig. 5c, d respectively. This actuator presented can open a new field on soft robotics with low voltage consumption and fast bending response.

Conclusions

In this paper, a multifunctional CNTF/DPMS actuator with large bending movement and the high generating temperature was fabricated and studied. Owing to CNTF high electrical conductivity and CTE of PDMS, the CNTF/PDMS actuator exhibited high bending angle of 324° and could reach a high temperature of 351 °C at a low driving voltage 8 V within few seconds. In addition, the CNTF/PDMS composite ETA exhibit stable movement and response in time after an applied cyclic On/Off voltage. Finally, the designed actuator showed great potential in soft robotics, artificial muscle, smart switchable devices that can work efficiently in extremely low temperatures.

Table 1 Bending angles comparison with

	L×l×s* (mm^3)	Voltage (V)	Actuation**	Time (s)	Recover (s)	Temp (°C)	References
GO/PI film	12×22×4	65	270°	8	19	~130	[12]
MWCNTs/WPU	48×10×0.37	7	28 mm	50	100	160	[8]
Agnws/PDMS		4.5	720°	40	60	~160	[7]
MWCNT/Al/Epoxy	16×5×1	6	10 mm				[5]
CNT/KAPTON		100	300°	10	30–50		[16]
CNT film/BOPP	70×18	5	360°	3	15	47	[17]
SACNT/PET	15×10×1	10	212°	0.55		92	[18]
SACNT/PET/BOPP	47×18	60	38.1 mm	~25	30	~67	[19]
Agnws/PP/PEDOT:PSS	45×9×2	7	360°	20		70	[20]
SACNS/PDMS	30×6×1	40	9.5 mm	<10	60	98	[21]
CNT/KAPTON	~40×10×2	30	12 mm	~23		~103	[22]
OUR WORK							
CNTF/PDMS	**20×5×1**	**8**	**324°**	**12**	**10**	**351**	

*Support information for actuator dimension

**Support information for bending angle and displacement comparison

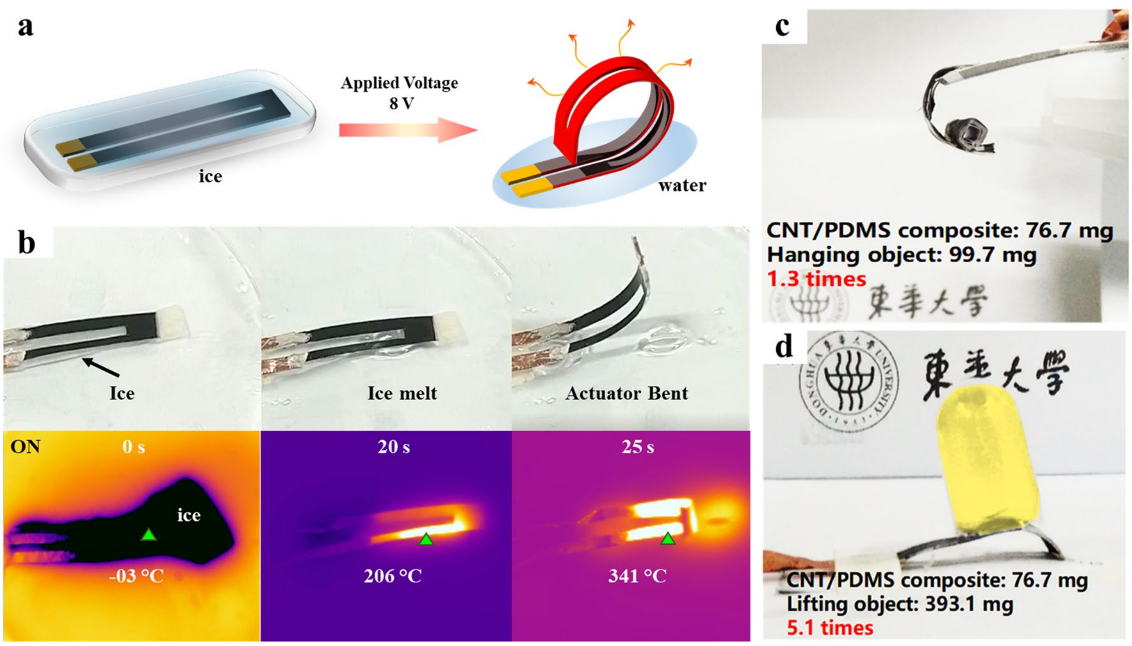

Fig. 5 Demonstration of CNTF/PDMS composite ETA in low temperatures and as soft robotic finger. **a** Schematic of the CNTF/PDMS composite ETA workability in cold environment with its **b** optical and thermal images under the applied voltage of 8 V. CNTF/PDMS composite ETA soft robotic finger that **c** grab and **d** lift objects

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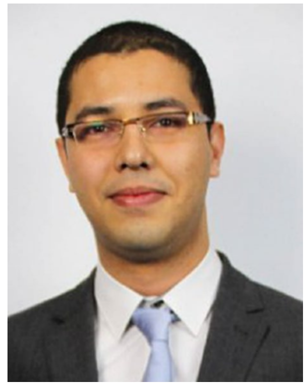

Mohamed Amine Aouraghe received his M.S. degree in textile engineering at ENSAIT (France) in 2012. In parallel, he received an M.S. research degree in textile manufacturing process from Lille 1 university (France) in the same year. Now his a Ph.D. student at Donghua university, college of textile (China). His main scientific research is focused on electronic textiles and multifunctional composites material based on carbon nanotube film.

Zhou Mengjie is a postgraduate student at the Donghua University. Her scientific research is focused on developing multifunctional soft actuator based on high conductive carbon nanotube film.

Yiping Qiu received his B.S. degree in Textile Engineering at Zhejiang Science and Technology University in 1982, M.S. degree in Textile Science at Auburn University in 1988, and Ph.D. degree in Fiber Science at Cornell University in 1992. He then did his post-doctoral training at Massachusetts Institute of Technology with Prof. Stanley Backer from 1992 – 1994. Afterwards, he worked as the Principal Materials Scientist in the Timberland Company, and an Assistant Professor at Kansas State University and North Carolina State University. In 2003 he joined College of Textiles at Donghua University as a University Titled Professor. He is the Executive Vice Chairman of Mainland China Region of the Society for Advancement of Material Processing and Engineering (SAMPE) and served as the Chairman of the Textile Engineering

Division, American Society for Mechanical Engineers. In 2014, he was awarded SAMPE Fellow. He was a technical consultant for five American Companies, including P&G, PPG, and Corning Inc. and a technical expert witness for three US law firms. He finished research projects from NSF, NIH, and NTC of USA. After back to China, Prof. Qiu has received funding from National High Technology Research and Development Program of China, National Defense Basic Science Research Program, and Shanghai Pujiang Program as well as projects from number of domestic and international companies. He is serving as an editorial board member for Textile Research Journal, Journal of Industrial Textiles, Journal of Adhesion Science and Technology, and Review of Adhesion and Adhesives. He has published more than 200 referred journal papers in peer-reviewed international journals and received one US patent and more than 100 Chinese patents. He has graduated more than 25 PhD students, and more than 70 MS students both in China and in the US.

Xu Fujun received his B.S. degree in Department of Applied Physics in College of Science of Donghua University in 2005, then obtained the joint cultivated Ph.D. of Donghua University and North Carolina State University in 2011. Now his Professor and Vice Dean of College of Textiles, Donghua University. His main scientific interests include: Carbon Nanotube fiber/ yarn/ composites, high performance fiber/ fabric/composites; 3D Woven Fabrics/Composites. In the above fields, Prof. Xu has more than 80 peer-reviewed papers published in the journals of Adv. Mater.; ACS Nano; ACS.AMI; Compos. Sci. Technol; and possesses 45 Chinese invented Patents. Now Prof. Xu serves as Senior member of the China Institute of Composite Materials (2017-2020), and Editorial board members of the "Composites Part B: Engineering"; "Journal of Textile Science & Fashion Technology"; "International Journal of Materials Science".