LARGE STROKE ACTUATION OF ALIGNED CNT-PARAFFIN COMPOSITE FILMS

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1. Introduction

Among active materials, paraffin wax is attractive for use in actuator devices due to its large stroke high and simultaneous large stress output [1]. High energy density [2, 3], and, depending on its thermal mass, relatively fast actuation. For example, at 150°C paraffin expands 20% under 200MPa of stress [2] and has an energy density similar to that of shape memory alloys.

Paraffin wax has previously been used in microscale pumps [4], valves [5], and actuators [6]; however, miniaturizing these designs requires multiple soft lithography steps to fabricate the necessary sealed microfluidic channels, reservoirs, and heaters to build the actuation system. The complexity of integration of paraffin microactuators has perhaps prevented their wider adoption as elements of micro-scale active material systems.

Moreover, it is challenging to integrate paraffin as an actuator because confinement of the paraffin is necessary to extract its volume change into useful and directed motion. Recently, Lima et al. confined paraffin wax inside carbon nanotube (CNT) yarns. The strong wetting of CNTs by paraffin prevented it from leaking out of the CNT network upon heating, instead causing the CNT yarn to expand radially. The yarn's twisted configuration coupled the radial expansion to a 8% contraction in length [7].

However, to our knowledge, micro-scale actuation using CNT-paraffin composites has not been explored. We previously showed that the interaction between anisotropic mechanical properties of CNT forest microstructures and an isotropic active polymer such as a hydrogel can enable anisotropic motion upon water exposure [8]. Here we demonstrate a high-stroke film actuator using paraffin-infiltrated aligned CNT “forests”, which are pre-compressed to enable expansion of paraffin to cause directed expansion of the nanocomposite film. The CNT-paraffin films expand reversibly by 20% in the vertical direction upon heating to 175°C, and this performance is repeatable over several cycles.

2. Fabrication

First, CNT forests were grown using chemical vapor deposition (CVD) from a thin film catalyst (1nm Fe supported on 10nm Al2O3) deposited by sputtering on a SiO2/Si wafer. The catalyst-coated substrate was annealed in a 400:100 sccm H2:He gas mixture at 775°C for 20 minutes, and then exposed to a 400:100:100 sccm H2:He:C6H6 mixture for 2-3 minutes for CNT growth. Rapid cooling of the CNT forest in the growth atmosphere is crucial for strong adhesion to the substrate, which prevents the detachment of the film the substrate during subsequent processing.

Schematics of the fabrication process along with the experimental setup used for both sample fabrication and mechanical characterization are shown in Fig. 1. To fabricate a CNT-paraffin film, a CNT forest (attached to the growth substrate) was placed on a hot plate at 100°C and compressed by a desired amount (typically 10-50% of the initial height) using a vertical precision actuator. While maintaining this temperature, molten paraffin wax was added to the silicon substrate, and promptly wicked into the CNT forest, which was constrained from expanding vertically upon infiltration. Constant load was applied during cooling. Last, the sample was placed on a spin coater and exposed to 75mW/cm² of ultra violet (UV) light for 35 seconds prior to spin coating. Excess paraffin was then spun away at 3000 rpm for 30 seconds while the UV light was maintained. The light is absorbed by the CNTs and
converted to heat, causing melting of the paraffin prior to and during spin coating.

All sample fabrication and indentation experiments were performed using a single axis precision actuator with both PID position and load control, which is driven using a voice coil actuator. The motion was measured using a linear optical encoder, while the load was measured using a S-beam load cell (Futek).

3. Results and discussion

Fig. 2a shows a mirrored optical image of the CNT-paraffin actuator side view at 25°C and extended by 17% at 150°C. The CNT-paraffin actuators were tested by optically monitoring the position of the top surface during heating. Three consecutive cycles of a CNT-paraffin actuator under no load are shown in Fig. 2b. The thermal strain increases rapidly after the paraffin melts, and then increases linearly as the wax undergoes linear thermal expansion after melting. The expected melting point for the paraffin used in these samples is 53-57°C. Some thermal expansion is also observed before the onset of melting. There is good agreement between the thermally generated strain over the three cycles with the exception of the initial heating.

The isotropic thermal expansion of the paraffin combined with the anisotropic CNT mechanical properties results in anisotropic thermal expansion along the film thickness. At elevated temperatures, the strong surface energy of the molten paraffin wax prevents it from flowing out of the CNT forest and instead the pressure of the molten wax does work against the stiffness of the forest, causing the material to expand. Strong adhesion between the CNT forest and its substrate prevents the forest from expanding laterally and from shearing from its support substrate. As a result, the motion of the actuator, powered by the volumetric expansion of the melting paraffin wax, is restricted to an extension along the CNTs.
Fig. 2. (a) Mirrored images of actuator cross section at 25°C (left) and at 150°C (right), showing a strain of 17%. (b) Thermal strain of CNT-paraffin actuator versus temperature, for three identical sequential heat-cool cycles.

Scanning electron microscope (SEM) images of a CNT forest crushed to 50% of its original height before paraffin infiltration are shown in Fig. 3a and Fig. 3b (close-up). The characteristic wavelike buckling of vertically compressed CNTs near the substrate is observed, as previously shown by Cao et al. [9] and others. However, unlike Cao et al. the buckling in the base of the forest is not recovered when the load is removed, because the induced elastic deformation energy is overcome by the van der Walls forces between additional CNT contacts created during compression of the forest.

The forest expands vertically during infiltration, as shown in Fig. 3c. Fig. 3d shows a SEM image close-up of a previously buckled region significantly expanded in height after infiltration. We see paraffin has fully infiltrated, due to the contrast and charging of the SEM image. Forests that are allowed to expand during paraffin infiltration develop large cracks on their top surface upon cooling after spin coating instead of undergoing a retraction in height. The crack formation is caused by local capillary aggregation of CNTs as the excess paraffin is spun out of the forest. During heating, cracked actuators do not have significant vertical strain, instead the cracks were observed to shrink. Forests that were restrained during infiltration have significantly less paraffin content prior to spin-coating and do not crack upon subsequent cooling. Only forests that were constrained vertically during infiltration result in functional actuators.

The dependence of the thermally generated strain on the amount of forest compression before/during paraffin infiltration is shown in Fig. 4a. All of these CNT-paraffin actuators were tested by applying a constant load while in contact with the top surface and then heating. The thermal strain curves of both samples can be considered to have three regions: (1) low thermal strain below the melting point, (2) rapid expansion during melting, and (3) further expansion driven by the increased pressure of the molten paraffin wax. As previously mentioned, forests not constrained during infiltration do not extend vertically upon heating. A larger forest compression results in larger thermally generated strain, but a smaller overall linear extension. The thermal strain curve for the forest compressed by 10% deviates significantly from the linear thermal expansion of pure paraffin and that of other shown actuators at elevated temperatures. This is due to the forest reaching full recovery to its pre-compressed height at about 150°C.

Thermal strain curves for a sample under constant load are shown in Fig 4b. Maximum strains of 19% and 12% are reached at 120°C under 0.1kPa and 1.0kPa respectively. Surprisingly, the strains reached under load surpass those reached in the no load condition. We hypothesize this is due to run-to-run variation in the density of CNT forests [10], which influences their mechanical properties and therefore influences the actuator performance. We expect an increase in density to correspond to a larger actuator stiffness at the expense of lowered maximum strain.

The actuators tested under constant load as low as 0.1 kPa exhibit a much more gradual expansion after the wax melting point, as compared to the actuator under no load. Actuators under load exhibit a slower and smaller thermal expansion before the
wax melting point and a larger expansion afterwards. This is further evident in the thermal strain curve under 1kPa of pressure. Under these conditions the actuator expands linearly up to 45°C, then begins to retract until 58°C, and finally collapses rapidly to zero strain. This behavior is explained by a rapid reduction in stiffness at 60°C once the wax is completely melted. The expansion after this is solely due to the pressure inside the wax increasing. Our measurements of the CNT-paraffin actuators show similar characteristics as the standard pressure-volume-temperature data for paraffin wax having similar molecular weight [2] to that used in our experiments (Fig. 4c). Because our actuator geometry couples the volumetric expansion of paraffin wax to a linear expansion in film thickness, the thermally generated strain of CNT-paraffin composite films can be directly compared to the volume change from the standard pressure-volume-temperature data.

Fig. 3. SEM image of crushed forest (a) and close up of buckled region (b). SEM image of CNT-paraffin composite (c), which was not constrained during infiltration and close up of buckled region (d).
Fig. 2. (a) Comparison of the impact of pre-infiltration forest compression to the thermally generated strain. (b) Thermally generated strain under constant load for a CNT-paraffin actuator compressed by 50% prior to infiltration. (c) Standard pressure-volume-temperature curve for a similar pure paraffin taken from [2].

4. Conclusion

We have fabricated and tested CNT-paraffin film actuators, exhibiting up to 20% thermal strain at 175°C. This high-stroke vertical actuation is enabled by strong capillary interaction between paraffin and CNTs, and engineering of the vertical compliance by compression of the CNT forest prior to and during paraffin infiltration. We propose two mechanistic regimes of the CNT-paraffin actuator: a high stiffness, low strain mode below the melting point and a low stiffness, high strain mode above the melting point.

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References


