Stacking nematic elastomers for artificial muscle applications

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Received 3 March 2006; received in revised form 10 April 2006; accepted 18 April 2006
Available online 10 July 2006

Abstract

Nematic liquid crystal elastomers are thermally actuated to produce macroscopic, anisotropic shape changes. Uniaxial contraction and extension of liquid crystal elastomers can be achieved by cycling the temperature of the material through the nematic to isotropic phase transition. In this report, a new approach is introduced by layering liquid crystal elastomer films to create thermally actuated stacks. A heating element and thermally conductive grease embedded between elastomer films provide a means for rapid internal heat application and distribution when a current is passed through the heating element, thus providing contractile force production in a minimal amount of time. Upon voltage application, stacks composed of two 100 μm-thick films and a single heating element produce 18% strain between contracted and relaxed states. In addition, the stacked elastomer films are capable of producing 10% contraction within 1 s and the blocked stress of the thermally actuated stacked films is calculated to be 130 kPa. The stacking approach provides new opportunities to use liquid crystal elastomers in applications requiring forces greater than those capable of being produced by single elastomer films.

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Keywords: Liquid crystal elastomer; Mechanical properties; Thermal actuator; Contraction forces; Stacked actuators

1. Introduction

The development of smart materials that respond to external stimuli and result in a change in the shape or size of that material is an ever developing technology. These materials, which include hydrogels, conducting polymers, dielectric elastomers, carbon nanotube films, and nematic or ferroelectric liquid-crystalline elastomers have been developed to respond to external stimuli [1–7]. The idea of using liquid crystal elastomers (LCEs) as an actuating device, i.e. an artificial muscle, was first suggested decades ago by de Gennes [8]. In particular, uniqueness of nematic LCEs stems from the fact that they exhibit reversible macroscopic and anisotropic contraction as the material is heated and cooled through the nematic to isotropic phase transition temperature, \( T_{NI} \) [9]. The macroscopic uniaxial contraction occurs parallel to the nematic director and arises from ordered mesogenic units that are anisotropically incorporated into a polymerized and cross-linked network. The development of an optimized nematic liquid crystal elastomer depends on tight coupling and orientation of the mesogenic unit to the polymer backbone. Neutron scattering has revealed that the orientational order of the mesogenic groups is coupled to the orientational order of the polymer backbone [10,11]. The orientation of the mesogenic units with respect to the polymer backbone will partly dictate the response of the material to application of an external field. It has been demonstrated that the origin of the anisotropic, macroscopic strain response is due to the loss of side-chain mesogenic order as the material passes from the nematic to isotropic phase [7,12]. The loss of mesogenic order is accompanied by a change in the conformation of the acrylate backbone which results in macroscopic contraction of the film.

A key application issue in the use of actuators, including nematic LCEs, is the ability to produce sufficient force for particular applications given the blocked stress of the material. The solution is to either chemically manipulate the material to increase its strength or increase the cross-sectional area of the material. Chemical alteration of a nematic elastomer by increasing the cross-linking within the network will strengthen the material but the strain response is sacrificed, making an inef-
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ective actuator. Therefore, the other approach is to increase the cross-sectional area of the elastomer.

Previously, our group has reported the physical properties of thermally actuated liquid crystal elastomers in the form of films and fibers [6,7,13–15]. Liquid crystal elastomers are produced as thin films in order to maintain the nematic phase alignment of the LCE through the bulk thickness of the material [7]. Alignment of the nematic elastomer originates with a chemical layer at the LC interface that has been mechanically rubbed. As the thickness of a sample is increased, the alignment through the bulk material is lost, limiting the thickness of an individual LCE film. As an alternative, the cross-sectional area of a LCE can be increased by stacking films. With other materials, such as electrostrictive polymers, increased force production has been achieved by stacking or rolling sheets of material onto itself with electrodes on either side of the polymer [16]. This concept led to the idea of stacking the LCE thermal actuators, but exposed the issue of efficient heat transfer into a stack. The solution is to maximize the surface area of a stack of films in contact with a heat source, which has been demonstrated in an LCE polymer bender [17].

Actuation of a stack could be induced two ways: external heating in a temperature controlled environment or inserting heating elements in a stack to heat the LCE films internally. External heating requires that a stack be in an insulated and strictly temperature controlled environment. In addition, heating would be dependent upon the thermal conductivity of the films, i.e. the ability for heat to efficiently transfer through the individual films in order to maintain uniform contraction of a stack. Previous studies have reported electrically activated Joule heating of liquid crystal elastomers by embedding a conductive material such as graphite fibers or powder into films [18,19]. In the current study, a unique approach is described that involves internal heating of an LCE stack, whereby individual LCE films are layered between thermal grease and heating elements. The embedded heating elements in combination with a thermal grease results in rapid heat distribution through an LCE stack allowing contractile force production in a minimal amount of time.

2. Experimental

2.1. Film production

Elastomers were created with a single nematogen and two cross-linking agents as previously described [6]. The structures of these materials are schematically shown in Fig. 1. Elastomers used for stacking contained 86 mol% of the monomer, 10 mol% of cross-linker A, and 4 mol% of cross-linker B.

The procedure of preparing elastomer films has also been previously described [7]. Briefly, mixtures of monomer, cross-linker, and 0.1 mol% of the photo-initiator Irgacure-369 (Ciba Specialty Chemicals) were heated above TNI and filled into a glass cell on a temperature-controlled hot stage. Glass cells were composed of rubbed poly-vinyl alcohol (PVA, Aldrich) coated glass plates, assembled anti-parallel and separated by Mylar spacers (Dupont), which determined the thickness of the LCE films (either 50 or 100 μm). To create LCE films, the mixture was cooled and aligned in the nematic phase and exposed to UV light for 8 min (6 mW/cm²). Films were removed from the glass cell by dissolving the PVA in 80 °C water. Alignment of the films was confirmed by viewing the films through crossed polarizers on a microscope stage. The physical properties of individual films have been reported previously and produce uniaxial contraction of ~20% strain with a TNI of 65 °C and a blocked stress of up to ~250 kPa [6].

2.2. Stack preparation

LCE films were cut into multiple pieces of the same size and coated with a thin layer of the thermally conductive compound Arctic Silver 5 (Arctic Silver, Inc.). Typical LCE film measurements were 2 cm × 1 cm (L × W). The thermal grease was applied by placing a small amount near the edge of a microscope slide (to provide a uniform straight edge). The slide was drawn across the film several times to produce a smooth, even layer of thermal grease. The thickness of the thermal compound layer was typically less than 100 μm.

Heating elements were designed to fit between LCE films in order to create an internal heat source for the stacks. Ni–Cr alloy (90% nickel, 10% chromium) wire was used due to its superior thermal conductivity and common use in high temperature resistance applications. A wire diameter of 50.8 μm (0.002 in.) was used to provide the best heating while remaining embedded within the thermal grease and not significantly reducing the uniaxial strain upon actuation. A zig-zag patterning of the wire was produced by using a breadboard with evenly spaced pegs to prefabricate the heating element. This procedure provided a uniform patterned heating element to cover the maximum surface area of the LCE films. Two Ni–Cr wire heating elements are shown in Fig. 2(a). The heating elements were placed in between films to create an LCE stack, schematically shown in Fig. 2(b). Typical stack dimensions were 2.0 cm × 1.0 cm × 0.03 cm (L × W × T).
Stacks were composed of either 50 or 100 μm-thick films and the width of the stacks varied between 0.5 and 2.0 cm for all samples tested. The forces produced by the stack contraction were recorded and saved to data files via MATLAB®.

3. Results and discussion

The response of an actuator is controlled by three principle parameters: the strain of the material, the blocked stress, and the speed of the response [21]. In order to quantify the strain and contraction rate, stacks were held under minimal load and subjected to heating and cooling cycles. As shown in Fig. 3(a), the stack was suspended with two glass holders and a standard binder clip to clamp the ends of the stack. Heating was accomplished by applying a predetermined current through the heating elements, which dictated the rate of contraction. Upon current application, a rapid initial increase of the contraction of the stacks was followed by a slower, steady increase to maximal strain. As shown in Fig. 3(b), when 40 mA was applied to stacks composed of two 50 μm-thick films the contraction measured at 1 s was 4.9 ± 0.3% and 14 ± 1% at 4 s following current application. The initial contraction rate increased to 9.0 ± 0.8% within 1 s when the active current was increased to 60 mA, while the strain measured 4 s after current application remained at 14 ± 1% (Fig. 3(b)). Application of currents higher than 60 mA led to degradation and discoloration of the stack. Interestingly, the only significant difference in the contraction rate of the stacks composed of 50 μm-thick films actuated at different currents was the initial contraction observed within 1 s. This difference is primarily due to the amount of heat introduced into the system upon actuation; a higher current adds more heat to the stack system. After this initial period, the contraction at different currents was nearly identical. This response would indicate that, following the initial difference in contraction, a threshold heating limit has been reached and the stack composed of two 50 μm-thick films is absorbing heat and contracting at a maximal rate.

The contraction rate and strain of stacks composed of two 100 μm-thick films were also tested. The active current applied to the stacks ranged from 70 to 150 mA. Testing the strain over a range of currents revealed a maximal contraction of ~18% between 100 and 120 mA. Lower currents inhibited full contraction of the films while higher currents led to film degradation. Two representative strain curves for stacks actuated under 80 and 100 mA are shown in Fig. 3(c). Stacks composed of 100 μm-thick films demonstrate 3–7% contraction within 1 s of the thermal stimulus. The strain achieved at each current consistently approached 15–20% but with differing rates, as opposed to the stacks composed of 50 μm-thick films (Fig. 3(b)). The different contraction rate for stacks with 100 μm-thick films is indicative of a limit in the rate of heat transfer through the elastomer films. While the thermal paste covers the entire surface of the film, the heat produced by the Ni–Cr alloy element must transfer across the thickness of the film itself in order to uniformly contract. Doubling the thickness of the films from 50 to 100 μm has doubled the thickness over which the heat must transfer, leading to a systematic decrease in the rate of contraction upon application of a current through the heating element.
The strain values observed for stacks composed of 100 μm-thick films were confirmation that the maximal strain of the LCE as a solitary film (20%) was being maintained when films were stacked with thermal grease and heating elements. Actuation of the stacks under optimized current conditions resulted in 18% strain between contracted and relaxed states over a minimum of eight cycles of 30 s heating and 30 s cooling. Within 5–10 s during these cycles the LCE stack had contracted to 80% of the full contraction. Stacks had ~10% strain between contracted and relaxed states when a 10 s hold time was used for both heating and cooling for at least eight cycles.

The force production of stacked films was examined independent of the contraction rate on a force monitoring apparatus, as shown in Fig. 4(a). Several film stacks were tested to examine the repeatability of the force production as a function of the cross-sectional area. Under isometric conditions, force measurements represent the sustainable force exerted by a contracted LCE stack. As shown in Fig. 4(b), a stack was mounted onto holders in the force monitoring device. In order to determine the maximum sustained force of a particular stack, the distance between the holders was initially less than the length of the mounted stack (Fig. 4(b)). In such a configuration, the film was...
initially free to contract upon application of a current. As contraction progressed, the stack was reduced until the stack began to exert a contractile force on the holders, which was monitored by a load cell aligned with the adjustable holder (Fig. 4(a)). In successive runs, the distance between the two holders was incrementally increased, reducing the slack during contraction and increasing the force exerted on the load cell. Fig. 5(a) shows the force profile of a stack composed of four 100 µm-thick films as the displacement between the stack holders was incrementally increased each run. In this stack, three heating elements connected in parallel were used to apply the thermal stimulus. The maximum sustained force prior to failure of the stack was 0.45 N. Ultimately the stack yielded when the force produced by the stacks was overcome by the force required to keep the holders stationary. Note that the equilibrium force produced by the stack increased over a time frame much longer than the times reported for experiments examining the contraction rates. In the force monitoring device, stacks were held at room temperature prior to application of a current with no resting current being passed through the heating elements.

The relationship between the cross-sectional area and the maximum force production was investigated for several stacks containing either 50 or 100 µm-thick films of varying widths. If the integrity of single films within the stacked LCEs is maintained, it is expected that the maximum force and cross-sectional area of the stacks should collapse onto a single curve. As shown in Fig. 5(b), this expectation is realized and the average blocked stress of this particular stack was 104 Pa. (b) Summary of the force production as a function of the cross-sectional area of the films within stacks. The key indicates the number of films (blue, red, or green) and thickness of films (circles or triangles) for each data point. The dotted lines represent the average maximum stress (black) ±20% (gray) produced by the stacks. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)

4. Conclusion

It has been demonstrated that thermally actuated LCE films can be stacked into multi-layered units for tunable force produc- tion over a 10-fold range. The incorporation of heating elements provides a unique way to internally heat a stack of LCEs to induce uni-axial contraction upon application of a current. The thermal grease serves a two-fold purpose of inhibiting film degradation in areas in contact with the heating element and distributing the heat over the surface area of the film. The method of stacking eliminates the need for an externally controlled environment for applications ranging from robotics, microfluidics, shape changing membranes, deformable fins for autonomous underwater vehicles, etc. Future studies include examining threshold contraction rates and force production as a function of the number of films in a stack and designing ways to incorporate the stacked films into mechanical devices.

Acknowledgments

The authors thank David Cylinder from the Princeton Plasma Physics Laboratory for helpful discussions. This work was funded by the Office of Naval Research. C.M. Spillmann acknowledges the National Research Council for a post-doctoral associateship.

References

Biographies

Christopher M. Spillmann is a research physicist at the Center for Bio/Molecular Science and Engineering, Naval Research Laboratory (NRL), Washington, DC. He received his BA in physics from the College of Wooster in 1997 and his PhD in biophysics from the University of Rochester in 2004. In 2004, he was awarded a National Research Council Associateship at the NRL. He has expertise in the field of cellular mechanics, receptor/ligand interactions and liquid crystal applications. His recent work with liquid crystals includes actuators and functionally active nano-materials.

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Banahalli R. Ratna is currently the Head of the Laboratory for Interfacial Interactions at the Center for Bio/Molecular Science and Engineering, NRL, Washington, DC. She received her PhD in physics from Mysore University, India. Her research interests include physics and application of liquid crystal and directed colloidal self-assembly. She has more than 140 research publications and patents. Since she joined the NRL in 1994, she has studied various electro-optic properties of liquid crystals, such as electroclinic and pyroelectric effects. Her interests also include the development of novel bio-based nanostructures. Currently, she is leading a group engaged in developing liquid crystal elastomers for artificial muscle applications and active nano-materials.