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Tuning the physical properties of a nematic liquid crystal elastomer actuator

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In this report we demonstrate the ability to tune the physical properties of a liquid crystal elastomer (LCE) by varying the amount and type of crosslinking within the elastomer network. LCE films composed of a single mesogenic compound were capable of uniaxial contraction when thermally actuated through the nematic to isotropic phase of the material. We probed the physical properties of the LCE films while varying the amount and concentration of two crosslinking agents and measured actuation strains of 10–35%, elastic moduli of 3–14 MPa, and transition temperatures ranging between 75 and 60°C. The viscous losses of the elastomers and the estimated work capable of being produced by the films were also evaluated. The ability to tune the physical properties of the LCE films allows for a wide range of applications including robotics, microelectromechanical systems (MEMS), shape-changing membranes, and/or microfluidics.

1. Introduction

A developing technology is the creation of smart materials that respond to external stimuli resulting in a change in the shape or size of that material. These materials include hydrogels, conducting polymers, dielectric elastomers, carbon nanotube films, and nematic or ferroelectric liquid crystalline elastomers [1–7]. They have been developed to respond to external stimuli, such as temperature, electric field, ion concentration, changes in pH, etc., and have a wide range of physical properties including stress, strain, and response time. The obvious attraction of these actuators is their use in applications where reproducible contractions and/or deformations are desired. The response of an actuator is controlled by three principal parameters: the contraction length (3strain) of the material, the stress it is capable of exerting, and the speed of the response. The optimization of these three parameters is important for successful implementation in a wide range of applications including robotics, microfluidics, microelectromechanical systems (MEMS), and shape-changing membranes.

A unique application of liquid crystals is the development of crosslinked polymeric liquid crystals with anisotropic properties that are able to mimic

biological materials such as skeletal muscles [7–9]. The supramolecular ordered assembly of liquid crystals provides the framework to incorporate anisotropy in response to an external stimulus. Polymerization and crosslinking of mesogenic units in an oriented nematic phase creates an ordered and anisotropic network. Thermoelastic studies of such materials led to the observation of several phenomena including memory effects, i.e. reversible contraction and relaxation through the phase transition of the elastomer resulting in anisotropic, macroscopic shape changes [10].

The development of a nematic liquid crystal elastomer (LCE) with an optimal thermostriuctive response depends on the coupling and orientation of the mesogenic unit to the polymer backbone. Previous reports have used neutron scattering to demonstrate that the orientational order of the mesogenic groups is coupled to the orientational order of the polymer backbone. For example, laterally attached mesogens within an elastomer exhibit large backbone anisotropy [9, 11]. Mesogens with lateral orientation (with respect to the polymer backbone) reduce the effect of soft elasticity, a phenomenon thought partly to arise from reorientation of the nematic director within the elastomer [12]. Coupling between the nematogen and the polymer backbone should be optimized to allow changes in mesogen orientation that are tightly associated with the orientation of the polymer backbone [13, 14]. The orientation of the mesogenic units with respect

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to the polymer backbone will partly dictate the response of the material to application of an external field. In terms of the change in molecular orientation upon thermal actuation, our laboratory and others have previously 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, 14]. The loss of mesogenic order is accompanied by a change in the conformation of the acrylate backbone, as revealed by macroscopic contraction of the film. The macroscopic contraction and change in the polymer backbone conformation is reversible due to the elastomer network created by the crosslinking agents [10].

Previously, we have developed and reported the physical properties of thermally actuated liquid crystal elastomers in the form of films and fibres [5, 7, 15, 16]. In each case a single crosslinking agent was used to produce the elastomers. In this paper we probe the effect of two different types of crosslinking agent on the mechanical properties of the resulting elastomer. By varying the concentration of the two crosslinkers we are able to tune the physical properties of a nematic side chain LCE film to optimize the strain, work capacity, and transition temperature of the films.

2. Experimental

2.1. Materials

All reagents and solvents were purchased from Aldrich and were used as received. The monomer C411U8, shown in figure 1, was synthesized partly using procedures described by Gray *et al.* [17] and adapted with a polymerizable acrylate group. The structure of the material consists of a rigid core composed of three phenyl rings with hydrocarbon chains attached to either end of the rings. The four-hydrocarbon spacer between the core of the molecule and the acrylate polymerization site was used to couple the orientation of the molecule tightly to the orientation of the polymer backbone.

The tri-functional disk-like crosslinker TAC-4 possesses three acrylate sites that can be involved in the polymerization process (figure 1). A similar crosslinker had been synthesized and used by Kim and Finkelmann [18]. TAC-4 was prepared by a standard esterification of trimesic acid trichloride and 4-hydroxybutyl acrylate. The second crosslinking agent used in this study was hexanediol acrylate (HDA), a commercially available fluid-like material that contains two sites available for crosslinking (figure 1).

Preparative chromatographic purifications were performed by employing flash chromatography on E. Merck 40–63 μm normal phase silica gel. Analytical

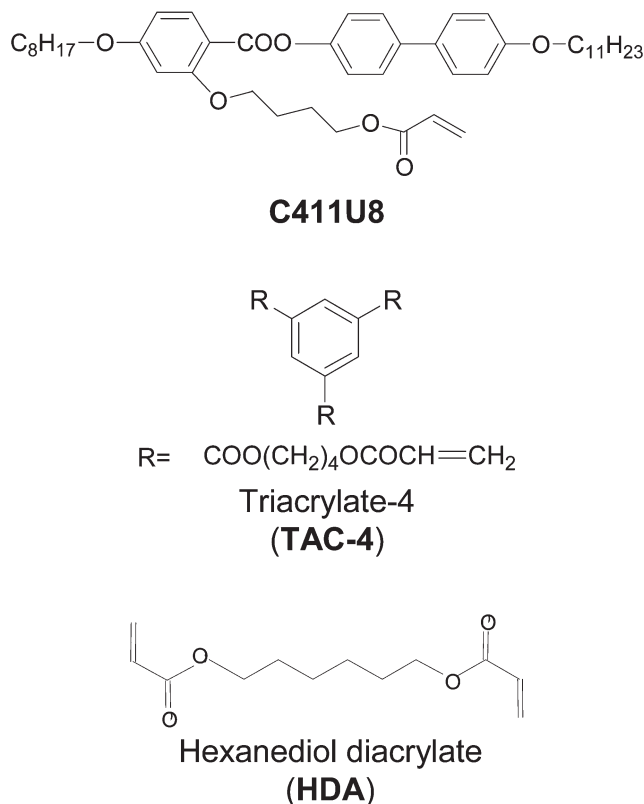


Figure 1. Chemical structures of the components in the nematic elastomer film. The materials include the monomer C411U8 (heating: K 50 N 74 I; cooling: Cr<0 N 74 I, °C) and the two crosslinking agents, TAC-4 and HDA.

thin layer chromatography was performed on glass silica plates (0.25 mm thick, E. Merck silica gel 60-F254). GC/FID sample analysis was carried out on a HP 6890 instrument equipped with a 30 m DB-5 column, with an oven program of 50°C (1 min) to 300°C (10 min) at a rate of 8°C min⁻¹. 400 Mhz ¹H NMR spectra were recorded on a Bruker MSL300 spectrometer in CDCl₃ or DMSO solutions. Differential scanning calorimetry (DSC) revealed C411U8 to have a large temperature range in the nematic phase, making it an ideal candidate for use in the LCEs.

2.2. LCE films

The preparation of elastomer films has been previously described [7]. Mixtures of monomer and crosslinker were dissolved in dichloromethane with 0.1 mol% of the photoinitiator Irgacure-369. Following evaporation of the solvent, the mixture was heated above T_{NI} and filled into a glass cell on a temperature-controlled hot stage. Glass cells were made of two antiparallel rubbed polyvinyl alcohol (PVA)-coated glass plates separated by Mylar spacers. The spacers determined the thickness of

the LCE films, which were either 50 or 100 μm thick. Once the cell was filled, the mixture was slowly cooled below T_{NI} ($\sim 50^\circ\text{C}$) and aligned in the nematic phase. The mixture was exposed to a UV light source for 8 min at $\sim 6\text{ mW cm}^{-2}$ to create the LCE film. Films were removed from the cell by dissolving the PVA in water at 80°C until the glass separated from the film. Alignment of the films was confirmed by viewing them through crossed polarizers on a microscope stage. The thermal transitions of the materials were determined using DSC (scan rate 5°C min^{-1}).

2.3. Mechanical studies

All mechanical measurements were performed on a TA Instruments Dynamic Mechanical Analyser (DMA) 2980. Films were held in a tension clamp consisting of a fixed upper clamp and a moveable lower clamp positioned on an air bearing. Three types of experiment were performed on the DMA: thermoelastic, isostrain, and stress/strain. Thermoelastic experiments were performed by ramping the temperature of the film $0.5^\circ\text{C min}^{-1}$ through the transition temperature of the material. The transition temperature for each thermoelastic cycle was calculated by dividing each cycle into the heating and cooling segments and calculating the numerical derivative. Plots of the numerical derivative were then fitted to a 3-parameter Lorentzian curve. Isostrain experiments were performed with a ramping rate of 1°C min^{-1} . Isothermal experiments monitored the strain on a film in the nematic phase (40°C) as a function of the force per cross-sectional area (stress). The force was ramped 0.001 N min^{-1} for stress/strain curves. Passive and active work loops maintained the same force ramping rate. Dynamic work loops had a temperature ramping rate of 1°C min^{-1} .

3. Results and discussion

The tensile strength and resistance of an LCE to applied forces are primarily dependent on the type(s) and density of crosslinker added in combination with the liquid crystal monomer. As a first step toward the creation of a nematic elastomer, the nematogen C411U8 was combined with the crosslinker TAC-4 at varying mole percentages. LCE films made with increasing amounts of TAC-4 (5, 10, or 15 mol%) led to several changes in the C411U8 films. The elastic modulus increased from 3 to 14 MPa with the 3-fold increase in TAC-4, which corresponds to an increase in the stiffness of the films. In films made with increasing amounts of TAC-4, the transition temperature from the nematic to isotropic state, T_{NI} , decreased from 75 to 65°C and the strain decreased from 35% to 7% in thermoelastic

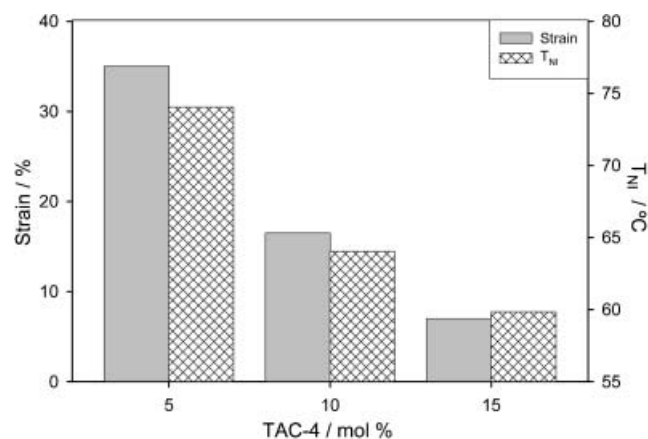


Figure 2. Thermoelastic effect of increasing mole percentage of TAC-4 from 5 to 15% in the nematic LCE films. The decrease in strain (grey) and the transition temperature (hatched) were the result of an increase in the crosslinking density within the LCE film.

experiments (figure 2). In addition, DSC revealed an increase in the glass transition of the material from 26°C with 5 mol% to 34°C with 15 mol% TAC-4.

Increasing the amount of TAC-4 in the elastomer also resulted in a significant decrease in the viscous losses of the LCE films. Passive work loops were used to measure the viscous losses of LCE films via cyclic scans of the stress versus the strain monitored under isothermal conditions in the nematic phase. The accompanying hysteresis of a cycle, figure 3(a), was determined by calculating the area enclosed by the increasing and decreasing stress curves. The area within the curve was then converted to mass specific work using a density of 1.04 g cm^{-3} for our films, based on several measurements of the volume and mass of film samples. All subsequent values of energy loss or active work of the films were calculated using the same density. Passive loops measure the history dependence of the material and provide a measure of the internal energy losses, i.e. energy dissipation, of the material. As shown in figure 3(b), there was a 50% decrease in the viscous losses of the film in the nematic phase when the amount of TAC-4 was increased from 5 to 15 mol% for passive work loops cycled between forces of 0.001 and 0.01 N. No significant difference in the energy dissipation was observed when the TAC-4 concentration was increased from 10 to 15 mol%. The cause of this apparent saturation was not determined, but may have been the result of some degree of phase separation of TAC-4 within the elastomer network. When viewed through crossed-polarizers under a microscope, the films crosslinked with increasing amounts of TAC-4 were observed with non-uniformities dispersed throughout the network.

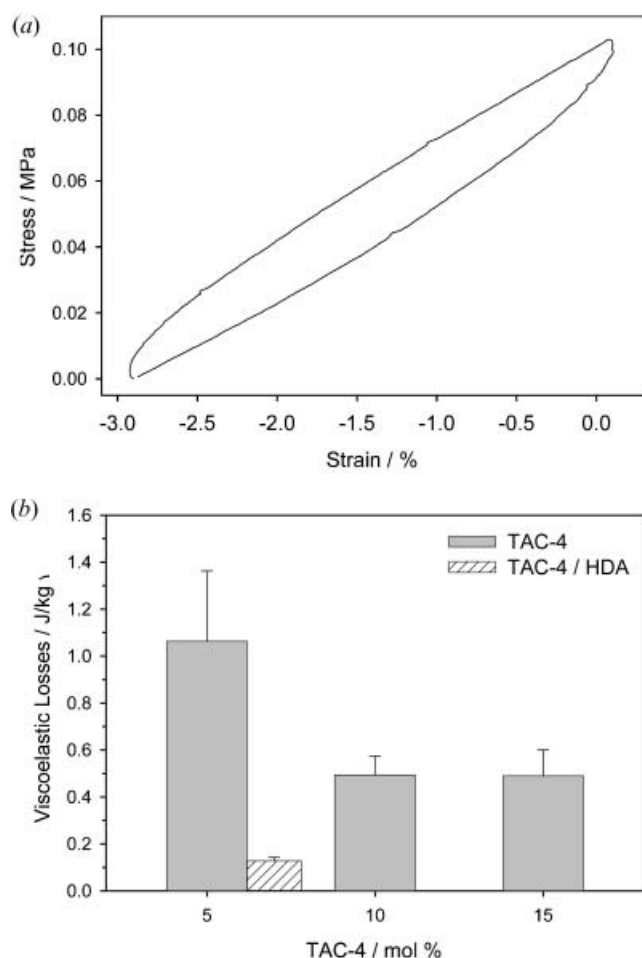


Figure 3. Viscous losses of C411U8/TAC-4 LCE films. (a) Passive work loop of an LCE film containing 10 mol% TAC-4. The viscous loss of the film in the nematic phase (40°C) was calculated to be 0.49 J kg^{-1} . (b) Summary of the viscous losses of films containing the nematogen C411U8 and the crosslinker TAC-4 (solid bars). For comparison, the hatched bar represents the viscous losses of films containing 5 mol% TAC-4 and 10 mol% HDA.

Just as the viscous losses of the films decreased with increasing TAC-4 content, a similar trend was observed for the work performed by the films. A work loop technique first described by Josephson [19], and subsequently used by others [20], was adapted based on the description given by Hebert *et al.* [21] to determine the work output of the LCE films. As shown in figure 4(a), the dynamic work loop employs components of both thermoelastic (segments 1 and 3) and isothermal (segments 2 and 4) experiments and operates in a counter-clockwise manner on a stress versus strain curve. In such a way, a closed loop is constructed and the area within the loop corresponds to an estimation of the work done by the film through an actuation cycle (assuming a film density of 1.04 g cm^{-3}). Dynamic work

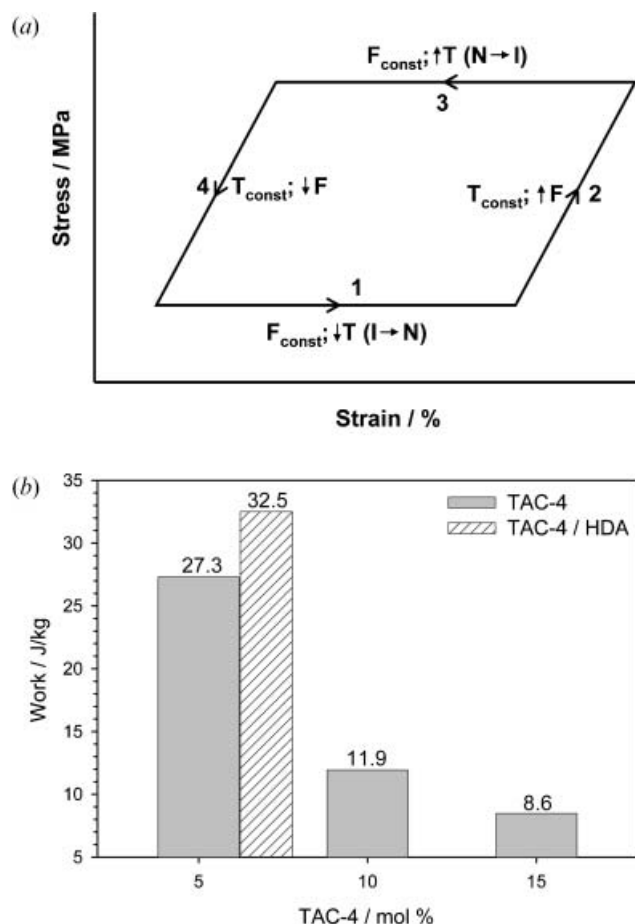


Figure 4. Dynamic work loop technique and work summary of LCE films with varying amounts of TAC-4. (a) Conceptual diagram of a dynamic work loop performed with LCE films. The elastomer film was first equilibrated in the isotropic phase under minimal load (0.002 N). The strain was monitored as the temperature was ramped to the nematic phase of the film (segment 1). Next, the force applied to the film was increased to a predetermined maximum (segment 2). In the third segment, the force was held constant as the temperature was increased at 1°C min^{-1} to the isotropic phase. Finally, under constant temperature the force was decreased and returned to the initial minimum load. (b) Work performed by LCE films as a function of TAC-4 concentration, showing a 70% decrease in the work with a three-fold increase in the amount of TAC-4 (solid bars). Films containing TAC-4/HDA had the greatest work capacity of all samples tested (hatched bar).

loops with TAC-4 LCE films were all conducted under the same conditions. The temperature was ramped between 40 and 75°C during segments 1 and 3 and the force ramped between 0.002 and 0.015 N through segments 2 and 4. The slow temperature and force ramping rates through a dynamic loop actuation cycle represent an upper limit of the estimated work performed by the films. As shown in figure 4(b), dynamic work loops on LCE films with increasing

concentrations of TAC-4 revealed a significant decrease in the amount of work performed by the film. The work decreased from 28 J kg^{-1} with 5 mol% to 9 J kg^{-1} with 15 mol% TAC-4.

From the three concentrations of TAC-4 used to crosslink the elastomer, C411U8 combined with 5 mol% TAC-4 produced the desirable qualities of high strain and a large capacity to perform work. However, a few drawbacks left room to improve the physical properties of the elastomers. For practical applications, a large energy input would be required to actuate the C411U8 films containing 5 mol% TAC-4 at the high transition temperature of 75°C . Ideally, a more desirable LCE would feature a low T_{NI} in order to reduce the energy required to switch the thermally-driven material. It is also useful and advantageous to have the ability to increase the elastic modulus and reduce the viscous losses while maintaining the largest possible strain and a large capacity to perform work.

As an alternative, a bi-functional crosslinker HDA was used to crosslink C411U8 polymer. The mechanical properties of the elastomer were studied to compare with the TAC-4 based elastomers. Stress/strain experiments on C411U8 combined with 10% HDA revealed an elastic modulus of 1 MPa, less than one-third of the elastic modulus of any film made with TAC-4 as the sole crosslinking agent. The low elastic modulus was expected and nearly matched the value of 0.9 MPa reported previously with nematic LCEs crosslinked with 10 mol% HDA [7]. Thermoelastic cycles revealed a strain of 38% through a transition temperature of 68°C , which is comparable to elastomers prepared with 5 mol% TAC-4 (see figure 2). This strain value is also similar to the strains observed in our previous study of nematogens crosslinked with 10 mol% HDA [7].

Both the energy dissipation and work capacity of the C411U8/HDA films were also determined. The viscous loss was found to be 0.23 J kg^{-1} , which was lower than the viscous losses of all films crosslinked only with TAC-4. The amount of active work performed by the film was 6.8 J kg^{-1} when stressed under 0.008 N of force. The application of ramping forces of 0.012 N or higher during the active work loop caused the HDA films to fail, emphasizing that the low work capacity of these films is primarily due to the low elastic modulus. Overall, LCE films crosslinked with HDA had positive qualities of high strain and low viscous losses. These advantages, however, were outweighed by the low elastic modulus and the small capacity to perform work. On the other hand, films made with TAC-4 had larger elastic moduli and a greater work capacity. Therefore, the approach was taken to tune the physical

properties of the C411U8 elastomer using a mixture of TAC-4 and HDA.

Films were made with 5 mol% TAC-4 and 10 mol% HDA as described previously. These films were subjected to heating and cooling cycles under constant load through the transition temperature. As shown in figure 5, the elastomer demonstrated 20–25% strain through T_{NI} when the temperature was ramped between 30 and 110°C . Within this range, 10 and 90% contraction was observed between approximately 45 and 85°C , respectively.

In conventional liquid crystals, there is an expected first order transition from the nematic to isotropic phase and a discontinuity in the orientational order of the material as a function of temperature. However, in the LCE films containing C411U8, TAC-4 and HDA we observe both a broad and smooth variation in the strain over a wide temperature range as the LCE undergoes a nematic–isotropic phase transition. There are at least two sources for this broad temperature range. First, multiple components added to a mixture result in a broadening of the phase transition temperature. Second, Selinger *et al.* have demonstrated that quenched disorder within the LCE network is a key limiting factor in a rapid phase transition [22]. In subsequent work, Selinger and Ratna employed a lattice model and concluded that both chemical heterogeneity and the distribution of local stresses within the elastomer network contribute to the observed broadening of the phase transition [23].

LCE films made with 10 mol% HDA and 5 mol% TAC-4 had a transition temperature of 65°C . This is a significant reduction from the transition temperature of

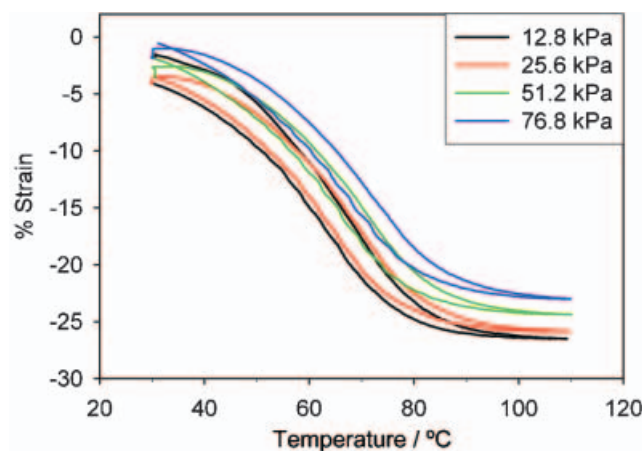


Figure 5. Thermoelastic plot of LCE film containing 10 mol% HDA and 5 mol% TAC-4. Cycles repeated through the same temperature range but under higher loads (held constant throughout a cycle) showed no significant change in the strain of the material.

75°C observed with 5 mol% TAC-4 and similar to the T_{NI} of LCEs made with 10 mol% HDA. The strain and transition temperature of the elastomer with two crosslinking agents were similar to the results obtained for the films containing 10 mol% TAC-4 (figure 2). The similarities may partly arise from the fact that 10 mol% of TAC-4, with its three polymerization sites, provides a similar number of crosslinking sites within the elastomer network as the combination of 10 mol% HDA and 5 mol% TAC-4. Isostrain experiments were performed on the LCE films to monitor the stress (force) as a function of the temperature as the film was heated through T_{NI} . The primary objective of isostrain experiments was to determine the stress necessary to prevent contraction of the film as it was slowly heated through the transition temperature, while held under constant strain. As shown in figure 6(a), when the film was held under 1% strain, the stress increased with increasing temperature until failure of the film at 250 kPa.

A representative stress/strain curve is shown in figure 6(b). The elastic modulus calculated at low stress values was 5 MPa. The elastic modulus of the LCE films crosslinked with both HDA and TAC-4 is slightly higher than LCEs crosslinked with 5 mol% TAC-4 and five times higher than the films crosslinked with 10 mol% HDA alone. The elastomer has been strengthened in an additive manner by combining the two crosslinking agents into the nematic elastomer.

The work output of an LCE partly depends on the ability of the elastomer to overcome the energy dissipation within the material through an actuation cycle. An example of a passive work loop performed on LCE films with 5 mol% TAC-4 and 10 mol% HDA is shown in figure 7. Multiple passive work loops performed on film samples at constant temperature resulted in negligible changes in the viscous losses. The calculated energy loss of 0.14 J kg^{-1} was almost an order of magnitude lower than for films containing only 5 mol% TAC-4, see figure 3(b), and 40% lower than the losses found in films crosslinked with 10 mol% HDA.

Based on the sites available for crosslinking within the elastomer network (three for TAC-4 and two for HDA), one might expect the energy losses for the HDA/TAC-4 films to be comparable to the losses for films with either 10 or 15 mol% TAC-4. However, we observed energy losses that were a third or less than for the 10 or 15 mol% TAC-4 films. Estimates of the crosslinking density based on the elastic modulus of the films do not account for the reduction in the viscous losses for the HDA/TAC-4 films [7]. A more reasonable explanation is the distribution of the crosslinker throughout the films. When the elastomer was crosslinked with 10 mol% HDA and 5 mol% TAC-4 there

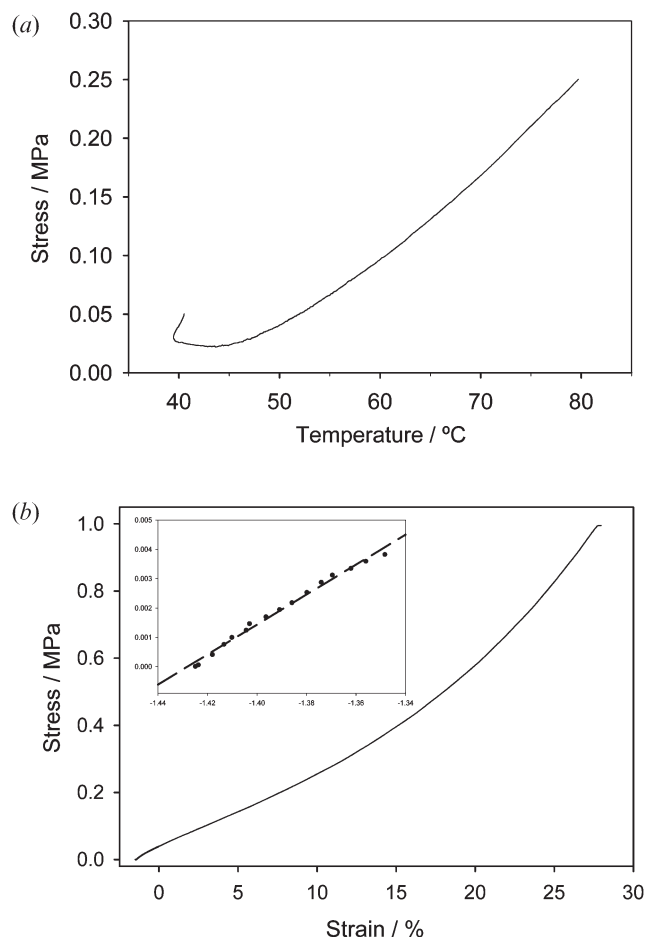


Figure 6. Isostrain and isothermal results for LCE films containing 10 mol% HDA and 5 mol% TAC-4. (a) The LCE film was actuated under 1% isostrain as the stress was monitored until failure of the film, providing a measure of the maximum sustainable stress by the film. (b) Isothermal curve performed on an LCE film in the nematic phase (40°C). The initial linear region of the curve (inset) provides a measure of the elastic modulus of the film, which measured 5.1 MPa for this particular sample.

were few, if any, non-uniformities compared with the films made with higher percentages of only TAC-4. The combination of 10 mol% HDA and 5 mol% TAC-4 in the elastomer network significantly reduced the viscous energy losses of the LCE films and optimized the elastic response compared with films crosslinked with only TAC-4.

Figure 8 displays three dynamic work loops for three different maximum ramping forces of 0.008, 0.012 or 0.016 N. The combination of TAC-4 and HDA in the elastomer network resulted in over a two-fold increase in the amount of work performed by the film compared to those crosslinked with HDA alone, when subjected to the same maximum ramping force of 0.008 N. The mass-specific work of the LCE films containing HDA

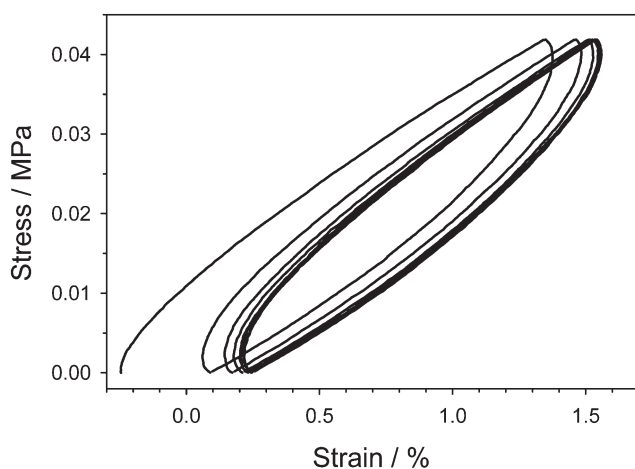


Figure 7. Example of passive work loops in the nematic phase (40°C) demonstrating the viscous loss of C411U8 LCE films with 5 mol% TAC-4 and 10 mol% HDA. The average energy loss calculated from this experiment was 0.14 J kg^{-1} .

and TAC-4 was slightly greater (15%) than films containing 5 mol% TAC-4, when compared at similar ramping forces, figure 4(b). In addition, there was over a two-fold increase in the work performed by the HDA/TAC-4 films when compared with films made with 10 mol% TAC-4. Taking into account the similar performance of these two films in thermoelastic experiments, the combination of HDA and TAC-4 has increased the amount of work performed by the LCE films. Also, the viscous loss of HDA/TAC-4 films was less than 0.5% of the mass-specific work, indicating that internal energy losses are minimal compared to the capacity of the films to perform work.

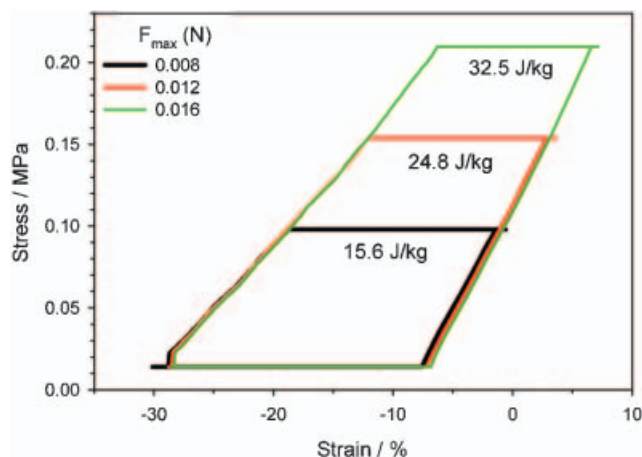


Figure 8. Dynamic work loops of LCE films containing 10 mol% HDA and 5 mol% TAC-4. F_{max} is the maximum ramping force applied and maintained on the film in the 2nd and 3rd segments of the loop, see figure 4(a). The work performed through each cycle is indicated within the loop.

In addition to the increased work capacity, HDA in combination with TAC-4 resulted in a transition temperature of 65°C . This value is a few degrees lower than the T_{NI} of films crosslinked with HDA but approximately 10°C lower than for films made with 5 mol% TAC-4, a concentration that produced the highest strain and greatest work when TAC-4 was the sole crosslinking agent. For practical application of thermally actuated LCE films, a lower transition temperature will increase the energy efficiency, i.e. less heat energy will have to be put into the system to actuate the material. If energy considerations are not important for a particular application, the films containing either 5 mol% TAC-4 or 10 mol% HDA provide a useful alternative, especially where high strains are important. However, the low elastic modulus of both these films, particularly the HDA films, will limit the capacity to perform work. In general, the combination of HDA and TAC-4 has optimized the physical properties of the films by increasing the work capacity, lowering the viscous losses, and reducing the transition temperature of the material.

This study has demonstrated the ability to tune the physical properties of an LCE by adjusting the type and amount of crosslinker used with a nematic material. A common challenge for many actuators is the balance between the strength of the material and the strain it is capable of producing. We have demonstrated the ability to adjust these parameters, thus providing a unique advantage to adapt the material properties to fit particular application requirements.

It is important to compare the response of our LCE films to other materials in order further to develop the response of the films toward specific applications. Skeletal muscle is able to perform many functions that require both gross and fine motor skills and is a useful standard for comparison. Assuming a blocked stress of 300 kPa, a strain of 25%, and the density of skeletal muscle to be 1.06 g cm^{-3} , the mass-specific work of skeletal muscle is $\sim 70\text{--}75\text{ J kg}^{-1}$ [24]. The maximum work we were able to perform with the C411U8 films crosslinked with both TAC-4 and HDA (32.5 J kg^{-1}) reveals that we have attained a mass-specific work of the same magnitude as that of skeletal muscle.

4. Conclusions

We have successfully created and tuned the physical properties of a liquid crystal elastomer film with the use of two different crosslinking agents. The nematic films contract with anisotropy in a reversible manner when heated/cooled through the transition temperature of the material. We recognize that the LCE films are limited by the efficiency with which heat can be transferred to

and through the film. Since actuation is temperature-controlled, it is apparent that the frequency of contraction is limited. Initial observations in our laboratory have revealed a maximum contraction (heating) frequency of the order of one Hertz and further studies are underway to evaluate this parameter. Current research focuses on developing similar liquid crystal materials with lower transition temperatures, and also on developing materials that respond to different external stimuli, such as electric fields.

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References

- [1] R.H. Baughman, C.X. Cui, A.A. Zakhidov, Z. Iqbal, J.N. Barisci, G.M. Spinks, G.G. Wallace, A. Mazzoldi, D. De Rossi, A.G. Rinzler, O. Jaschinski, S. Roth, M. Kertesz. *Science*, **284**, 1340 (1999).
- [2] M. Brehmer, R. Zentel, G. Wagenblast, K. Siemensmeyer. *Macromol. Chem. Phys.*, **195**, 1891 (1994).
- [3] A. Lendlein, A.M. Schmidt, R. Langer. *Proc. natl. Acad. Sci. USA*, **98**, 842 (2001).
- [4] Z.S. Liu, P. Calvert. *Adv. Mater.*, **12**, 288 (2000).
- [5] J. Naciri, A. Srinivasan, H. Jeon, N. Nikolov, P. Keller, B.R. Ratna. *Macromolecules*, **36**, 8499 (2003).
- [6] R. Pelrine, R. Kornbluh, G. Kofod. *Adv. Mater.*, **12**, 1223 (2000).
- [7] D.L. Thomsen, P. Keller, J. Naciri, R. Pink, H. Jeon, D. Shenoy, B.R. Ratna. *Macromolecules*, **34**, 5868 (2001).
- [8] P.G. de Gennes. *Cr. Acad. Sci. II B*, **324**, 343 (1997).
- [9] H. Finkelmann, W. Kaufhold, L. Noirez, A. Tenbosch, P. Sixou. *J. Phys. II*, **4**, 1363 (1994).
- [10] C.H. Legge, F.J. Davis, G.R. Mitchell. *J. Phys. II*, **1**, 1253 (1991).
- [11] P. Keller, B. Carvalho, J.P. Cotton, M. Lambert, F. Moussa, G. Pepy. *J. Phys. Lett. Paris*, **46**, 1065 (1985).
- [12] M. Warner, P. Bladon, E.M. Terentjev. *J. Phys. II*, **4**, 93 (1994).
- [13] B.R. Ratna, D.L. Thomsen, P. Keller. *SPIE Smart Struct. Mat.: EAPAD*, San Diego (2001).
- [14] W. Kaufhold, H. Finkelmann, H.R. Brand. *Makromol. Chem.*, **192**, 2555 (1991).
- [15] D.K. Shenoy, D.L. Thomsen, P. Keller, B.R. Ratna. *J. phys. Chem. B*, **107**, 13755 (2003).
- [16] D.K. Shenoy, D.L. Thomsen, A. Srinivasan, P. Keller, B.R. Ratna. *Sensor Actuat. A-Phys.*, **96**, 184 (2002).
- [17] G.W. Gray, J.S. Hill, D. Lacey. *Mol. Cryst. liq. Cryst.*, **197**, 43 (1991).
- [18] S.T. Kim, H. Finkelmann. *Macromol. rapid Commun.*, **22**, 429 (2001).
- [19] R.K. Josephson. *J. exp. Biol.*, **114**, 493 (1985).
- [20] R.J. Full, K. Meijer. *SPIE Smart Struct. Mat.: EAPAD*, San Diego (2000).
- [21] M. Hebert, R. Kant, P.G. de Gennes. *J. Phys. I Fr.*, 909 (1997).
- [22] J.V. Selinger, H.G. Jeon, B.R. Ratna. *Phys. Rev. Lett.*, **89**, 225701 (1–4) (2002).
- [23] J.V. Selinger, B.R. Ratna. *Phys. Rev. E*, **70**, 041707 (1–7) (2004).
- [24] C.J. Pennycuik. *Newton Rules Biology*, Oxford University Press, New York, p. 20 (1992).