SWOLLEN LIQUID CRYSTAL ELASTOMERS AS ARTIFICIAL MUSCLES

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ABSTRACT

We demonstrate, for the first time, the low voltage-driven electromechanical effects in liquid crystal elastomers (LCEs) swollen with low molecular weight liquid crystal (LMLC). Thin film polydomain (POLY) and monodomain (MONO) LCEs were embedded in a well-known LMLC, 4-n-pentyl-4-cyanobiphenyl (5CB), and introduced between transparent indium tin oxide electrodes with 100µm separation. Absorbing 5CB into POLY and MONO LCEs, shape changes were obtained in these materials by application of a small voltage. That is, compared to unswollen LCEs, a dramatic ~200 times decrease of the critical fields was found for electromechanical effect in swollen LCEs. The response time when the field was switched on in both POLY and MONO is below 2s. The results expect the outstanding potential of swollen LCEs as a low powered devices and actuator (e.g. artificial muscles).

Keywords: liquid crystal elastomers, electromechanical effects, artificial muscles.

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1. INTRODUCTION

Liquid crystal elastomers (LCEs) are interesting because of their large mechanical response, for example, length change as a function of temperature (Finkelmann et al., 1981; Brand and Finkelmann, 1998; Kupfer and Finkelmann, 1991). The macroscopic behavior of these materials arises from the coupling between the elastic properties of the cross-linked alkoxy polymer network and liquid crystalline degrees of freedom. The basic structure of LCE is shown in Fig. 1, it involves liquid crystalline ordering of mesogenic groups chemically tethered as side chains to a cross-linked polymer network.

The LCE material studied here were invented and developed by Finkelmann and co-workers (Finkelmann et al., 1981; Brand and Finkelmann, 1998; Kupfer and Finkelmann, 1991; Disch et al., 1994).

At first only polydomain sample could be produced for which there are domains with different director orientation, denoted by a unit pseudo-vector $\mathbf{n}$. But then it was discovered that applying a large enough strain could reorient the domains giving rise to a uniform director orientation (Brand and Finkelmann, 1998; Disch et al., 1994). Then monodomain samples, i.e. liquid single crystal elastomers (LSCEs), were successfully made using two cross-linking steps with the second cross-linking step performed on a stretched film. These films turned out to have a spatially uniform $\mathbf{n}$ parallel to the stretching direction.

![Fig. 1. LCE ingredients.](image-url)
Fig. 2. Polydomain (top) and monodomain (bottom) LCE samples. (a) In the nematic phase, the monodomain LCE is optically transparent, while POLY-LCE is opaque. (b) At high temperature above $T_{tr}$ ($> 80^\circ C$), both POLY and MONO-LCEs are transparent, while MONO-LCE exhibit drastic shape changes.

At low temperature, in the nematic phase, the polydomain is optically opaque because of the strong light scattering by random director orientations [Fig. 2(a)]. The monodomain is optically transparent because its birefringence axis or director orientation is uniform in the entire sample [Fig. 2(a)]; the strong light scattering on random domains is no longer present. In the isotropic phase both polydomain and monodomain sample are clear.

While monodomain LCEs exhibit a drastic length and shape change at the nematic-isotropic phase transition of the side chains [see Fig. 2(b)], the cross-linked polymer network without side chains has no such phase transition. The drastic length and shape changes of LSCEs at the nematic-isotropic phase transition led to the suggestion by de Gennes et al. that LSCEs may be useful components in artificial muscles (de Gennes et al., 1997; Hubert et al., 1997). A more suitable property for them may be a shape change by application of electric fields rather than temperature, i.e. an electromechanical effect. In current dry LCEs, however, very high electric fields are required to induce enough electromechanical effects (Cladis and Brand, unpublished).

The sensitive response of the director orientation on an electric field is another well-known cooperative effect in low molecular weight liquid crystals.
(LMWLCs) (de Gennes and Prost, 1993). Only a small voltage (~ 1 V) is required to change orientation of nematic LMWLCs giving a large electro-optic response.

Therefore it seems natural to combine the properties of LMWLCs with those of LCEs by swelling a LCE sample with LMWLCs to enhance the LCE response to the electric field so that the unusual shape changing properties of LCEs could lead to viable devices (see for example Cho (2000)), driven by small electric fields rather than temperature. Here, we check this idea by investigating the shape changes of LCEs swollen with LMWLCs in an AC electric field and, indeed, find evidence to support it. In the previous studies (Yusuf et al., 2003; Yusuf et al., 2004a; Yusuf et al., 2004b), we have investigated the swelling dynamics of LCEs swollen with LMWLCs.

II. EXPERIMENT

The polydomain (POLY) and monodomain (MONO) LCE films we prepare are ~ (25.0±1.0) μm thick and have an area of 300 μm × 50 μm. The samples are embedded in LMWLC for swelling between two transparent indium tin oxide (ITO) electrodes with very clean SiO surfaces as shown in Fig. 3. In the present study we use a well-known nematic LMWLC, 4-n-pentyl-4-cyanobiphenyl (SCB) that was homeotropically aligned. The cell gap was controlled by a polymer (Mylar) spacer of 100 μm in thickness.

Fig. 3. Picture of the LCE swollen with homeotropically oriented LMWLC. An alternating electric field (0, 0, E0) is applied to the sample cell.
In the electric field part of the experiments, an alternating electric field of the frequency \( f = 50 \, \text{Hz} \) with rectangular waveform is applied between the electrodes, i.e. \( E = (0, 0, E_z) \) at constant temperature \( T = 26 \, ^\circ \text{C} \) controlled to \( \pm 0.1 \, ^\circ \text{C} \) by a temperature regulator (CHINO Digital Controller DBS500). Measurements of the electro-mechanical effect were made using a polarizing optical microscope (Nikon E600W POL) on which a charge-coupled-device camera (SONY XC-75) was mounted. The mechanical deformations and the temporal changes of the electromechanical effect were captured by a computer.

**III. RESULTS AND DISCUSSION**

As described in our previous communications (Yusuf et al., 2004a), the equilibrium swollen state of POLY and MONO LCE samples swollen with LMWLC is achieved after about 2 hours. The observations of the shape changes were done in this state.

In Fig. 4, the contraction (displacement), \( \delta \), at the top of the edge of swollen POLY and MONO samples from their equilibrium swollen state, i.e. \( V = 0 \), as a function of applied voltage is shown. The temperature is fixed at 26 \( ^\circ \text{C} \). The solid lines are fit to

\[
\delta_{\text{POLY}}(V) = 20.87 - 22.18 \exp(-V/2.9),
\]

for the swollen POLY [Fig. 4(a)] and

\[
\delta_{\text{MONO}}(V) = 20.00 - 22.12 \exp(-V/3.4),
\]

for the swollen MONO [Fig. 4(b)], as a “guide to the eye”.

The displacement increase and saturates with a maximum value of about 21 \( \mu \text{m} \) in the swollen POLY and 20 \( \mu \text{m} \) in the swollen MONO samples. According to the fit, saturation is reached at about 10 \( V \) for both samples.

The insets in Fig. 4 clearly show a relatively small threshold filed, \( V_s \), for the onset of the observed electromechanical effect: \( V_s^{\text{POLY}} = 0.6 \, \text{V} \) for the swollen POLY [Fig. 4(a)] and \( V_s^{\text{MONO}} = 0.6 \, \text{V} \) for the swollen MONO [Fig. 4(b)]. In
dry LCEs, the displacement is arising for about ~ 1 V/μm [7, 13]. Therefore, by assuming that the threshold field for the swollen POLY and MONO are about 0.5 V/100 μm (100 μm is the sample thickness), it is 200 times smaller than that of dry LCEs.

Thus, the electromechanical effect of LCEs increases by swelling with LMWLC. The electric field acts predominantly on the LMWLC molecules which maintain their collective properties inside the LCE. They in turn entrain the tethered mesogenic side chains pulling at the cross-linked network.

Figure 5 shows optical micrograph for electromechanical effect observed in the shape changes (seen as displacement δ in Fig. 5) in the edge (the boundary of swollen MONO and homeotropically aligned 5CB) of swollen MONO sample.

![Image](https://via.placeholder.com/150)

Fig. 4. Voltage dependence of the displacement δ for swollen POLY (a) and MONO (b). Insets: expanded version of graph to show the threshold voltage for the LCE shape change for POLY (a) and MONO(b). T = 26 °C and f = 50 Hz.
Applying a voltage of 3 V to swollen MONO, the edge shrinks by about 10 μm. This shape change is because the LMWLC (5CB) molecules easily reorient parallel to the field, $E_0$. This cooperatively leads to the reorientation of many mesogenic side chains simultaneously which in turn changes the network shape making the sample thicker along $E$ and thinner perpendicular to $E$. Thus, the electromechanical effect is induced by the cooperative effect with LMWLC. The mesogenic side chains in swollen LCUs can effectively couple with each other via LMWLC molecules dispersed in the networks. Therefore a relative weak electric field can trigger a macroscopic shape change.

![Optical micrograph for the electromechanical effect observed in the shape changes](image)

Figure 6 shows the voltage dependence of the response times for swollen POLY and MONO samples when the electric field is switched on. The inverse of the response time ($1/\tau_m$) in swollen POLY and MONO are clearly proportional to $V^2$.

The solid lines in Fig. 6 are fit to

$$\frac{1}{\tau_m}^{\text{POLY}} = 0.51 + 0.10 \, V^2,$$

for swollen POLY [Fig. 6(a)] and

$$\frac{1}{\tau_m}^{\text{MONO}} = 0.34 + 0.06 \, V^2,$$

for swollen MONO [Fig. 6(b)]. This result is consistent with the response behavior of ordinary liquid crystals.
In the swollen MONO sample, the response time for the shape changes is slightly higher than that in the swollen POLY sample. This effect could be due to the fact that the rotation of LMWLC molecules and the mesogenic side chains is resisted by the elastic network. An effect that is not larger in MONO due to the stretching process during the LSCE preparation. The response time, $t_{res}$, with respect to the electric field is below 2 s for both swollen POLY and MONO samples.

IV. CONCLUSION

We studied the electromechanical effects of the POLY and MONO LCEs swollen with LMWLC and found that the threshold field for the electromechanical effects observed in the shape changes in both the swollen POLY and MONO LCEs is comparable to that of LMWLC and thus about two orders of magnitude smaller than that of dry LCEs. The inverse response time after switched on the external electric field for both swollen POLY and MONO
samples a quadratic dependence on the field strength. This is consistent with
the inverse response time for ordinary liquid crystals.

Because of this low voltage-driven electromechanical effect and speed
response, it is expected that swollen LCEs have outstanding potential for wide
applications as low powered devices and actuator (e.g. artificial muscles).

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