



Evaluation of the Important Properties of Liquid Crystal Elastomers (LCEs)

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Abstract

Liquid Crystal Elastomers (LCEs) have been the object of growing interest in recent years due to the combination of mesogenic ordering and rubber elasticity which allows them to respond to thermal stimuli by changing their shape, size and optical properties. The use of LCEs thus makes it possible to convert small quantities of external energy to mechanical energy which makes it easy to be used as energy harvesting. This paper will firstly focus on thermal and mechanical properties of LCEs it will be seen that they will be affected by elasticity of the polymer backbone and the nature of mesogenic monomer. Following this, the focus will turn to the electric field, light, and thermal energy that can be converted into mechanical energy induces thermomechanical, electromechanical and electrooptical effects by such materials. It will be observed that the shape of LCE materials can be changed through changing orientation of LC. Additionally, swollen samples can minimize threshold field by a factor of 200 compared with unswollen samples. Then, the emphasis will go to the impact of mixing nanoparticles with the LCE samples on its behaviour. This is done by increasing crosslinking agent content of the sample and then its impact is shown by the strain-stress curve which leads to reduction in the degree of nematic phase and increase in Young's modules. The paper then gives a future direction to expand the work for instance its application in photovoltaic systems to convert solar energy into electricity.

Introduction

Back to 1975, the de Gennes predicted an interesting property of liquid crystal elastomers [1]. It is found that the shape of liquid crystal elastomers change reversibly after applying an external stimulus, such as gradient of temperature, electric field or light due to the variations produced in its orientational order [1,2]. Since that time, LCEs have become a worldwide topic for researchers and industrial applications due to the wide range of possibilities that LCEs can be such as micromechanical systems (in atomic force microscopes, valves or artificial muscles), propulsion systems, smart surfaces or solar cells [2]. LCEs are significant materials since they combine both the orientational order of liquid crystals (mesogens) and the amorphous properties of soft materials (i.e. the rubber elasticity of polymer networks). This combination of properties permits variations in shape to be produced in a component. Such transformations are brought about via any process which changes the order of the mesogens which are cross-linked to the backbone of the elastomer [3,4].

The networked structure of LCEs limits the movement of the whole molecule; however the low crosslinking density allows relatively free movement of the mesogens. The netpoints of lightly cross-linked LCE are far from each other in the structure. Accordingly, movements of chain segments on a smaller length scale are comparatively independent from netpoint deformation. This allows mesogens inside the network freedom to set up liquid crystalline order which is independent on a microscopic length scale [4,5].

There are two basic domains of LCEs, and classification is dependent on the director orientation “n”. This is the average LC ordering direction of side chains composed of mesogenic groups. If the groups are macroscopically disordered, they are referred to as a polydomain sample, whereas groups which are macroscopically ordered in the LC state are referred to as a mono-domain sample [6,7].

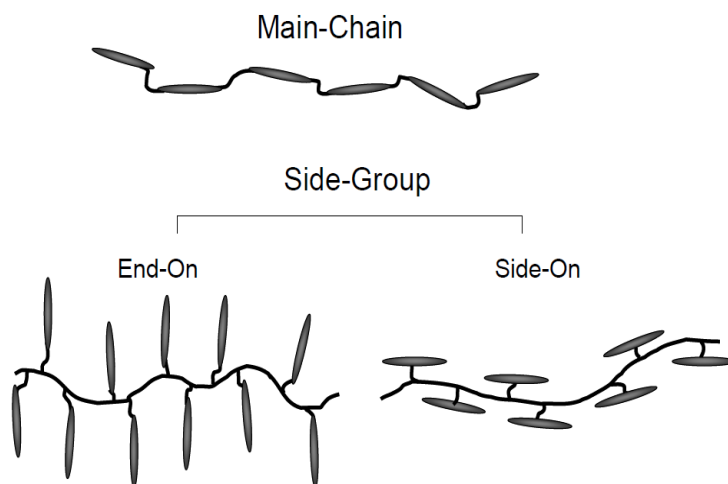


Fig.1: Schematic diagram of a main-chain LCEs and a side-chain LCEs [8].

A human body is composed of muscles which have an anisotropy structure and through contraction and elongation along the fibres axis can create work. It can be said that, the quantity of muscle’s work depends on three variables. They are strain through which the muscle can shorten, stress to exert and contract frequencies. The average values of these variables are 25 %, 350 kPa and 5–10 Hz, respectively [9]. Therefore, the capacity of modifying the shape with low strain, called soft elasticity, makes LCEs to be an attractive material in the creation of artificial muscles for robots.

A first-order phase transition from the nematic to the isotropic phase produces a large change in orientational order. Due to the presence of anisotropic intermolecular forces, liquid crystals demonstrate long-range order, an order which is not dependent on molecules possessing a permanent dipole moment [4]. As is shown in fig.2.

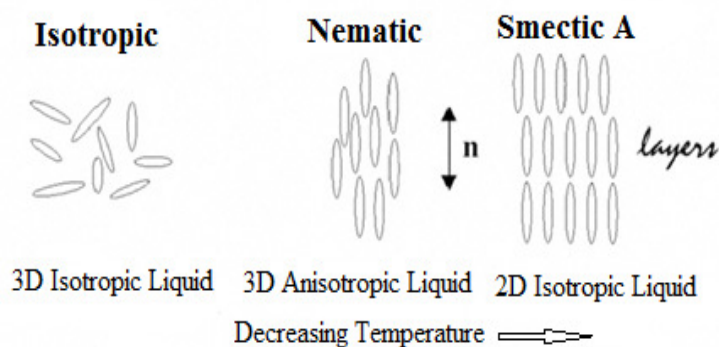


Fig.2: Shows isotropic, nematic, and smectic A diagram [10].

This paper designed to review some important properties of LCEs such as thermal and mechanical properties of LCEs including stress/strain. Then, it will be followed by addition of nanoparticles and crosslinking density impact on the properties of some samples.

Properties of LCEs:

The properties of LCEs differ from those of other materials since they combine the rubber-like elasticity of network structures and the orientational order of mesogenic groups.

A. Thermal Property of LCEs

Thermal property of LCEs can be affected by, among other factors, the overall flexibility of the polymer backbone, the nature of the mesogenic monomer, the spacer type and length, and both degree and type of crosslinking [10,11].

The flexibility of the backbone has an impact on the glass transition temperature (T_g) of LCEs. The T_g of acrylate or methacrylated-based LCEs is higher than that of siloxane-based elastomers, since the Si-O bond is longer than and can be rotated more easily than a C-C bond. Furthermore, Si-O has a larger bond angle than that of C-C, and since there are no extra substituents on the oxygen atom, rotation of an Si-O bond is easier than for a C-C bond. Generally, siloxane-based LCEs are good materials for use at room temperature [3,4].

The length of the spacer has an impact on the T_g of LCEs. In the case of acrylate-based side-chain LCEs, the rise in number of methylene units on the side chain from two to six causes a 30°C reduction in the T_g value for the material [4,12].

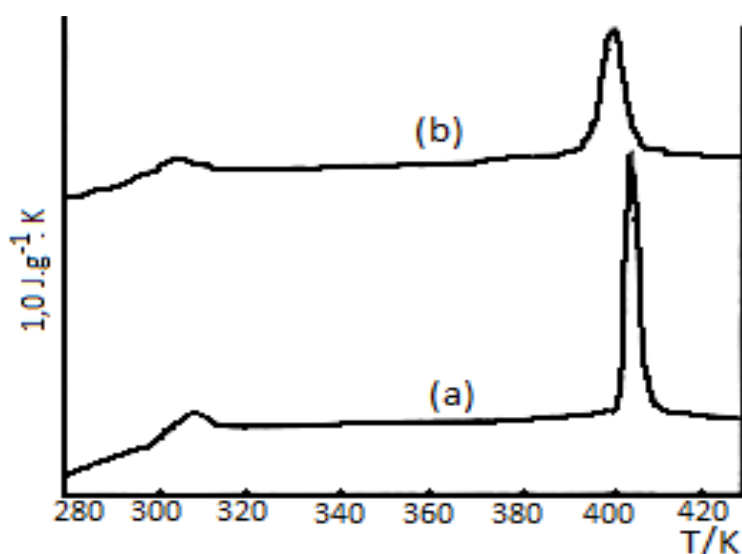


Fig.3: Shows (a) uncrosslinked main chain Liquid Crystalline Polymer (LCP) (b) crosslinked LCE, heating rate 20K/min [4].

As is shown in fig.3, a low crosslinking density causes a small variation ($\pm 5K$) in mesophase transition temperature, the magnitude of which depends on the nature of crosslinking agents and molecular-weight variation before and after crosslinking. Non-mesogenic crosslinking agents can drop the T_g , which affects the LC order, however T_g rises with the molecular weight increase that accompanies the crosslinking process [4,13].

B. Mechanical Properties of LCEs

In their initial condition, LCEs are in a polydomain state (a domain is a particular area which has its own favoured orientation) without special measures [7]. The many domains within a polydomain LCE have orientations misaligned with respect to one other, thus there is no regular global orientation in its mesophase.

Monodomain LCEs can be produced by aligning the separate domains, since the mesogenic groups of LCEs can be easily reoriented. In some Side Chain LCEs (SCLCEs), relative small elongation (20% strain) can reverse monodomain structures, while larger elongation is required to provide the same effect in Main Chain LCEs (MCLCEs) [4,7]. In this part the material acts similar to a conventional rubber. Section two of fig.4, shows that stress plateau corresponding-to-monodomain (P-M) transition. Further section three shows that, the stress starts to increase with the rise of the applied strain and the P-M transition is ended, while small extra orientation can be obtained.

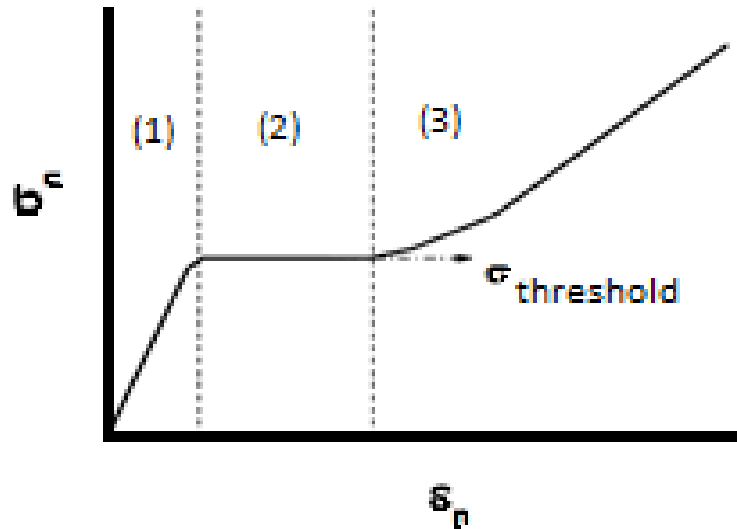


Fig.4: Shows stress-versus-nominal strain curve of LCEs.

LCE polydomains are usually opaque since the arbitrary director orientations of the domains produce scattering of incident light. However, by applying a large enough uniaxial extension, a sample of LCE will undergo polydomain-to-monodomain (P-M) transition and the material changes from opaque to transparent [14].

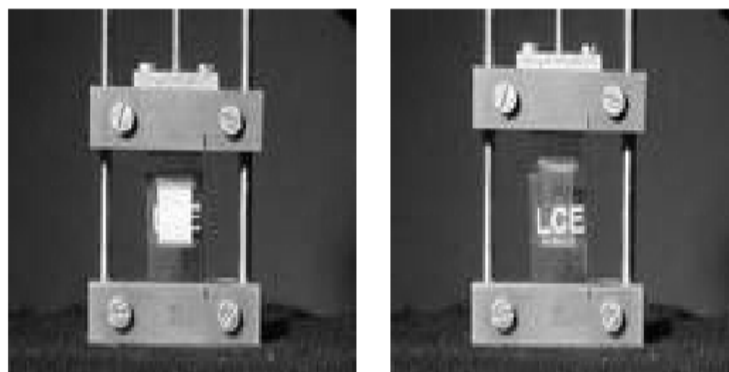


Fig.5: Originally opaque polydomain (left) becomes transparent monodomain (right) [4].

Transition occurs in P-M by reorientation of individual domains rather than by domain growth. Under uniaxial tension, the revolution of each domain leads to an overall change in orientation of the mesogens in a polydomain LCE fig.5 [4,14].

The polymer backbones within each domain orient along the local director in a polydomain LCE. In the whole network, the director of an individual domain misaligns in such a way that there is no ordering on a large-scale, since the domain size is roughly a few microns. A uniaxial extension applied to a polydomain

elastomer induces each domain to rotate such that local directors align along the loading axis. Eventually, the stretched sample changes from opaque to transparent [4].

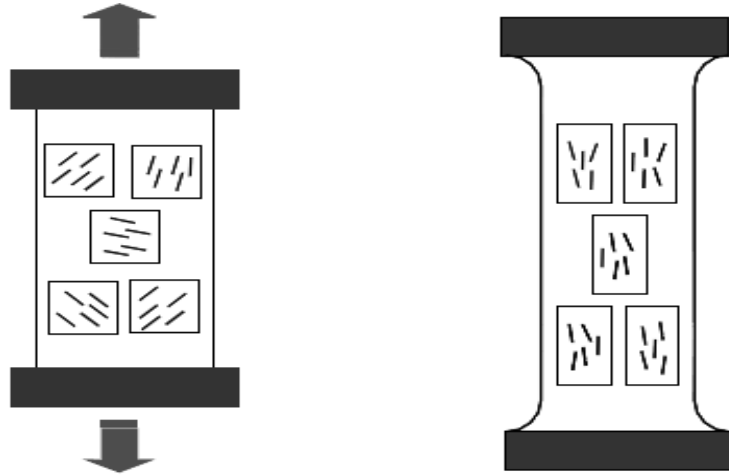


Fig.6: Representation of a stretched polydomain nematic elastomer undergoing a P-M transition by Terenjev [4].

The soft elastic-region is the P-M transition plateau referring to [4]. The stretching of the LCE network accompanies rotation of domains and can be obtained without a large stress response. Theoretically, ideal soft elasticity allows uniaxial deformation without resistance, but in practice, the elasticity of LCEs makes them good candidates for efficient damping materials and artificial muscle [15].

a) Effect of temperature on mechanical property:

Since LCEs demonstrate viscoelasticity, their mechanical properties will be affected by temperature and time. If temperature is increased, the state of the sample changes from the mesophase to the isotropic-phase, and the soft plateau area (P-M transition) starts to be reduced. The stress versus strain curves of cyclic siloxane crosslinker monomer and octasilaxane chain extender (C11(MeHQ)Si8XL10) for the synthesis of a polydomain main chain (LCE) at high temperatures is shown in fig.7 [4]:

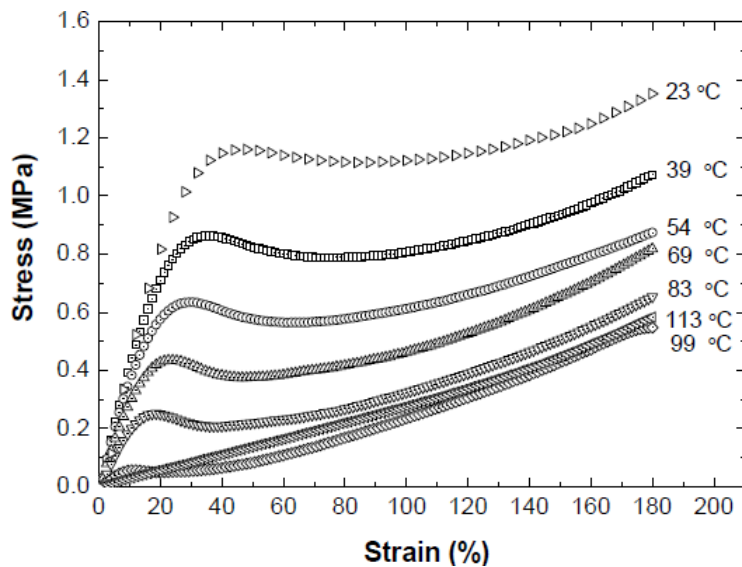


Fig.7: Nominal stress-strain curves of C11(MeHQ)Si8XL10 LCE 4b ($T_{cl} = 1.4^{\circ}C$) at elevated temperatures [4].

At room temperature the curve simply contains three regions, but as temperature raises, Young’s modulus, plateau threshold stress and plateau length are reduced. See table I.

Table.I: Threshold stress and Young’s modulus of C11(MeHQ)Si8 at elevated temperatures [4].

Temperature (°C)	Young’s modulus (MPa)	Threshold stress (MPa)
23	5.46	1.16
39	4.86	0.86
54	4.52	0.63
69	3.63	0.43
83	2.43	0.25
99	0.84	0.06
113	0.33	-

Above the mesophase to isotropic phase transition temperature (T_{lc-i}), LCEs are isotropic rubbers, while below T_{lc-i} they become anisotropic, having highly temperature and time dependent behaviour [4,16].

b) Impact of crosslinking density on the mechanical property:

In [4] five LCEs (4a-4e), C11(MeHQ)Si8X L5- C11(MeHQ)Si8XL25 were structured with different crosslinking agent content (XL) 5 mol % - 25 mol %. As is shown on the stress-strain curve (sketched at room temperature):

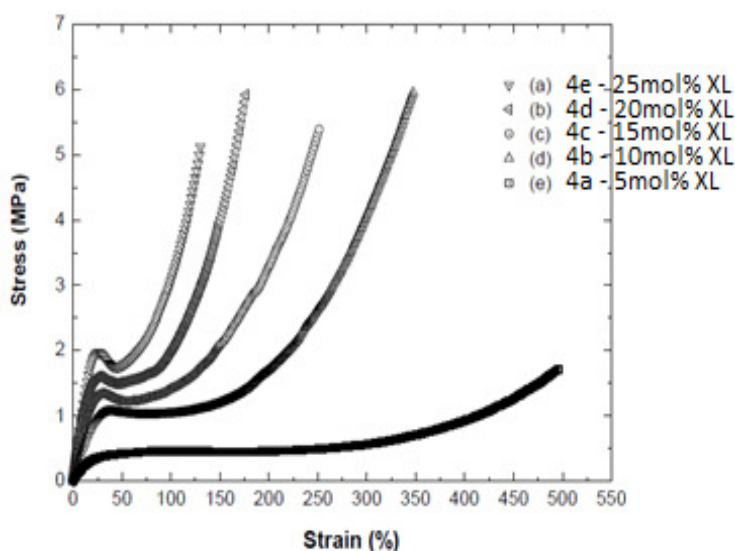


Fig.8: Effect of XL content on the mechanical properties [4].

It is concluded that with the rise in crosslinking agent content, there is a rise in the Young’s modulus and the threshold stress required to initiate P-M transition. Furthermore, an increase in crosslinking agent reduces the P-M transition plateau area and leads to a lower sample elongation at failure [4]. Should be well aligned

Table.II: Mechanical properties of LCEs 4a-4e [4].

	Young’s Modulus (MPa)	Threshold Stress/Yield Stress (MPa)	Elongation at break (%)
4a- 5 mol%XL	2.9	0.4	592
4b- 10 mol%XL	6.3	1.2	347
4c- 15 mol%XL	8.7	1.4	250
4d- 20 mol%XL	11.5	1.7	176
4e- 25 mol%XL	12.2	2.0	130

Where: XL is a crosslinking agent.

C. Photomechanical Property of LCEs

Irradiation with Ultra Volt (UV) light can be used to bring about photo-induced contractions/ expansions of LCEs integrated with azobenzene derivatives. This contraction/ expansion minimizes the order parameter which is the result of cis/trans-isomerisation. The rod-like trans-azo-benzene moieties stabilize LC alignment, while the bent cis constitute the minimum LC order parameter. This change of the polarization orientation will cause a gradual change of the LCE order and flexure [17]. A three-dimensional deformation (photo-induced bending) is obtained by utilizing nematic LCE films mixed with azobenzene before being expanded in suitable solvents or heated above their T_g values in air [18]. The restricting absorption of photons helps bending and shrinks the surface area [17], fig.9. A sample of such material floating on water moves away from light source because of deformations. The direction of movement is controlled by off-centre irradiation in an angle deformation of LCE [19].

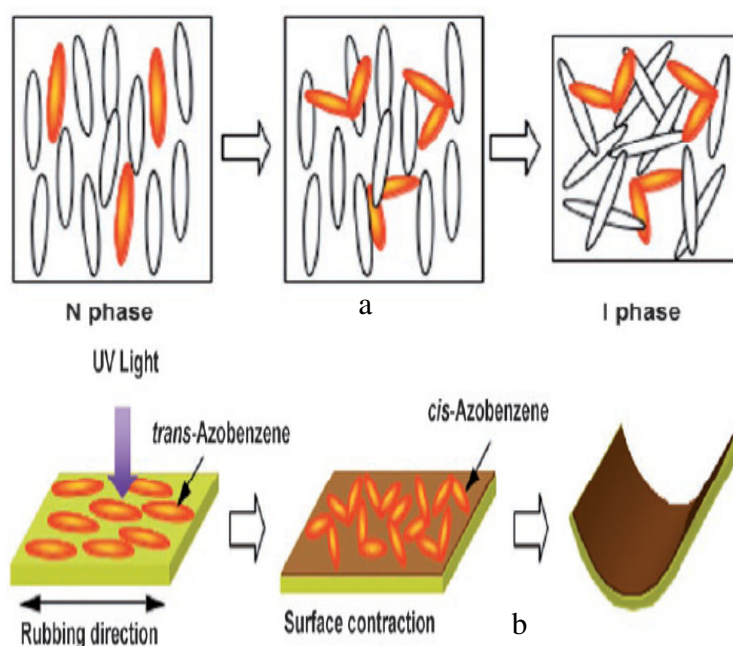


Fig.9: Shows (a) photoinduced contraction caused by photochemical phase transition (b) mechanism of the photoinduced bending behaviour [20].

Since in optimised conditions, the photochemical phase-transition for azobenzene -containing LC polymers can be induced during nanoseconds, these photo-responsive LCE materials are suitable for fabrication of high-speed actuators. Through use of these principles, light energy can be converted to mechanical power. These deformation processes do not require batteries or control systems since they are driven by light alone [17,21]. Compared with the two-dimensional case, three-dimensional deformation offers better results for artificial muscle applications, however the following limitations are relevant [15,18]:

- Slow response in the order of minutes/seconds.
- Bending is not efficient at room temperature.
- Low forces are induced upon photo-irradiation.

Some of these issues were mitigated when films of ferroelectric, azobenzene-containing LCE with a high order LC and low T_g were prepared by photo-polymerization under an electric field. Light with a wavelength of 366 nm bent the film at room temperature (with a response time of 500 ms) and the film was able to recover its initial state entirely when exposed to visible light. The force induced by photo-irradiation was 220 kPa, which is close to contraction force of human muscle (roughly 300 kPa.) This rapid and strong

mechanical response to light may increase the application of ferroelectric LCEs in artificial muscle for micro-opto-mechanical systems [18].

D. Electro-Mechanical Properties LCEs

LCE materials have the ability to change their shapes when electric field is applied. They produce an electromechanical response, reorienting their directors parallel to the electric field, an effect which is the basis of LC displays [7,12,20].

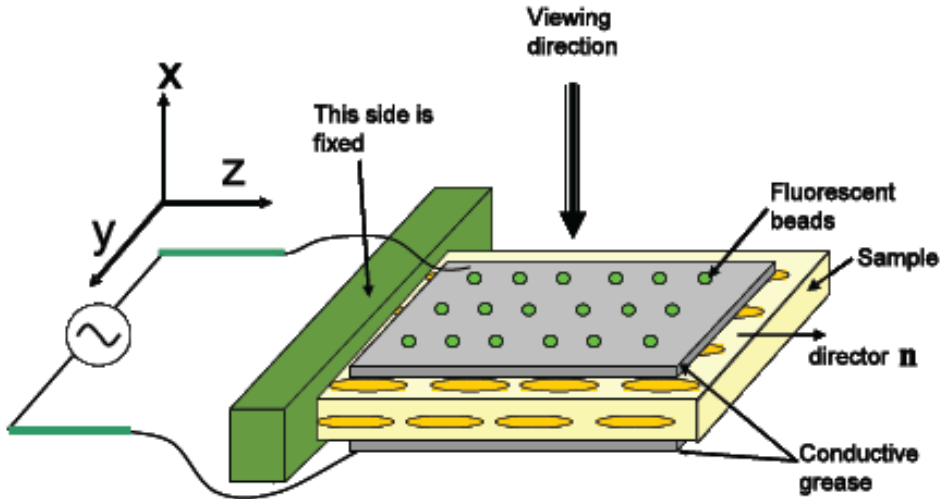


Fig.10: experimental setup for electrically induced deformation [22].

A sliced poly-domain sample with 20µm thickness was embedded in 4-cyano 4-pentylbiphenyl (5CB) (enlarged to 40µm after swelling) to investigate the electro-mechanical response. When a perpendicular field was applied to the electrodes, the sample shape varied with respect to its displacement from the balanced swelling state, see fig.10. The temperature was fixed at 26°C, and the solid line is given by

$$\delta(V) = 12.88 \left[1 - \exp\left(\frac{-V}{7.97}\right) \right] \quad (1)$$

With the rise of the voltage at constant frequency of 50 Hz, the sample shape changes further and saturates at 13µm [7].

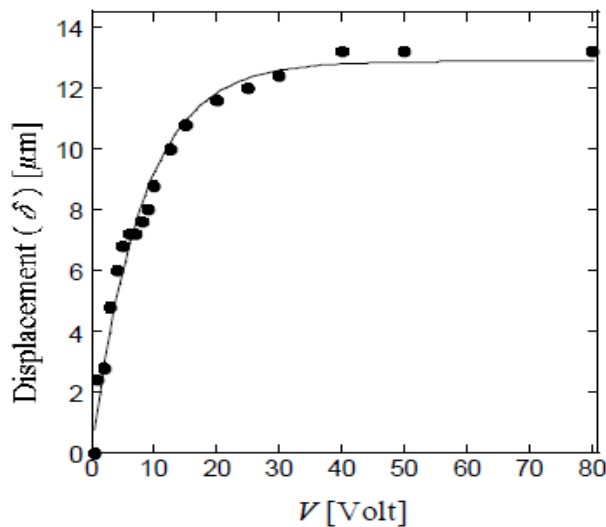


Fig.11: Displacement versus applied voltage at T=26°C [7].

To rotate the director in dry LCEs, a high electric field is required, whilst in swollen LCEs, a small electric field is sufficient to rotate the director and create a large change in sample shape [14]. Swollen nematic elastomers show macroscopic deformation, and their directors rotate 90° with application of a high electric field [23]. In references [20,24,25], a sample of 100 µm thickness had a field of 0-10 V applied, and a 1-20 µm shape change was recorded. It was found that in swollen samples, the threshold field was reduced by a factor of 200 when compared to unswollen samples.

Reference [14] studied the electromechanical and electro-optical impact of a material which had polydomain multifunctionality (MCLCE) and positive dielectric anisotropy having been swollen using 4-cyano 4-pentylbiphenyl (5CB). It was found that an electric-field almost instantly induced P-M transition within the swollen polydomain, followed by large shape change. Larger polydomain to monodomain transition changes were obtained than for side chain LCEs (studied in reference [24]). The use of ferroelectric LCs offers:

- High degree of mesogen ordering.
- Fast control of molecular alignment using an electric field, due to the availability of spontaneous polarization.

These data provide evidence that LCEs are attractive materials for fabrication of soft actuators, allowing large magnitude, high-speed deformation and either two-dimensional (contraction) or three-dimensional (bending) transformation [18].

E. Thermo-Mechanic Properties of LCEs

LCEs demonstrate a linear, anisotropic mechanical response to electrical and thermal stimulation, due to the ability of the LC director to change direction [26]. When temperature rises, the length of main-chain LCEs monotonically shrinks parallel to the director initiated by the ordering of mesogenic units. However, there is an expansion perpendicular to the director orientation, with quicker reduction in the vicinity of the apparent nematic-isotropic transition temperature, and then the sample returns to original shape after cooling [27].

With the rise of crosslinking agent the shrinking of the sample will increase. The length and size of the sample also varies during heating/cooling, causing hysteresis at the nematic-isotropic transition. The higher the sample cross-linking density, the lower the thermal fluctuation of the LC mesogenic main-chain molecules within the polymer system. LCE materials experience reversible macroscopic and anisotropic shrinkage when they are heated from the nematic-to-isotropic phase transition temperature and then cooled. Nematic LCEs are soft, elastic materials which have a stress/stain characteristic similar to that of human muscle [26].

Incorporating the nanoparticles can modify the elastic behaviour of the material. If the quantity of nanoparticles is high enough in a sample of LCE, the stiffness of the material will increase. The objective of adding nanoparticles to the LCEs is to enhance the response of the material to the external stimuli without changing its elastomer properties. A high concentration of nanoparticles can disturb the elastic network. However, embedding a low quantity can improve several interesting properties, such as dielectric anisotropy when the material is embedded with ferroelectric particles or introduces an electromechanical response when carbon nanotubes along the director are incorporated to LCEs [28].

Four samples were tested at different AuNPs concentrations in reference [26]. The LC director was measured, as was the elastomer strain as a function of the stress applied parallel to the long axis of the films. It is observed that with a rise in colloidal gold concentration (i.e. increased crosslinking-agent density), there was an increase in the Young's modulus and stiffness of the elastomer material. Both the type and concentration of the crosslinking agent play an important role in the resistance of LCEs to applied stress [26].

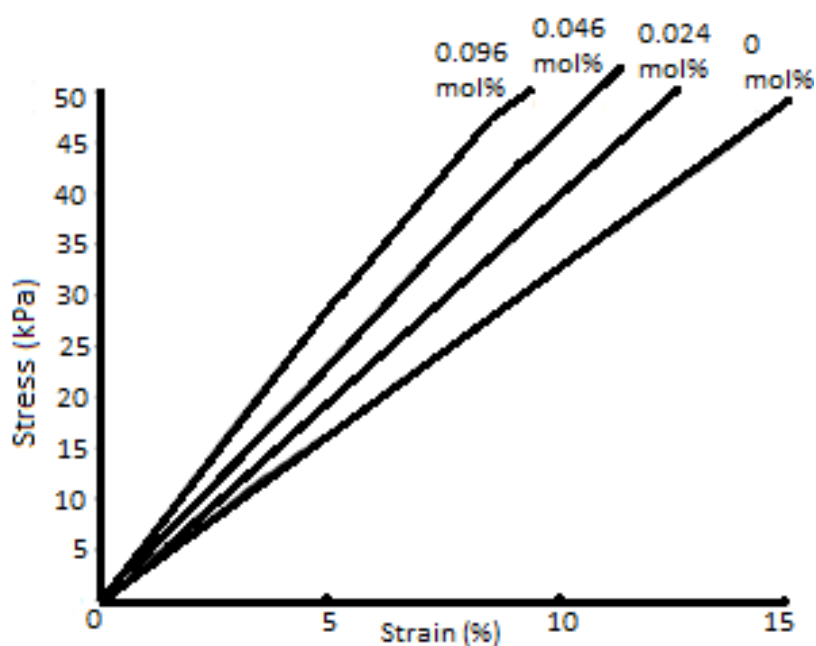


Fig.12: Stress-strain at different AuNP concentrations [26].

Following this, the thermoelastic behaviour of LCEs at various AuNPs concentrations was tested by heating and cooling a sample. The range was between 50-130 °C, with a ramp rate of 0.5 °C/min (across the nematic to isotropic transition region), with strain being plotted as a function of temperature at a constant static force [26].

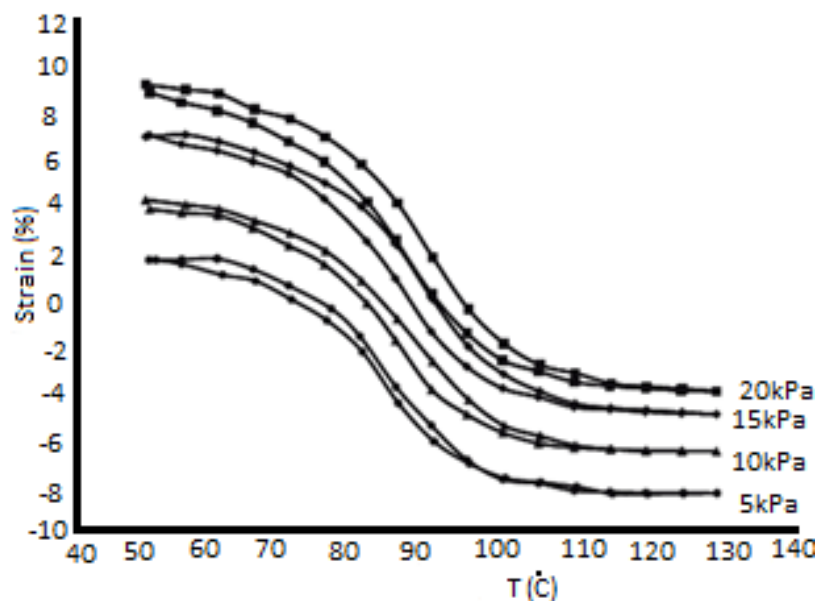


Fig.13: Thermoelastic curve of sample containing 0.048 mol % of AuNPs when different preloads (5-40 kPa) are applied [26].

The rate of change in mechanical strain with respect to time ($\Delta\epsilon$ %/min) was measured for the elastomers at various temperatures, in order to ensure that embedded AuNPs raise the thermal conductivity of the material. An initial heating ramp of 20°C/min was applied, at which the strain rate of the elastomer sample remains constant, following which the rate was increased to 50 °C/min. It was found that samples containing AuNPs demonstrated a higher change in strain rate, since they possess greater thermal conductivity characteristics. These characteristics improve the heat transfer to the elastomer material [26].

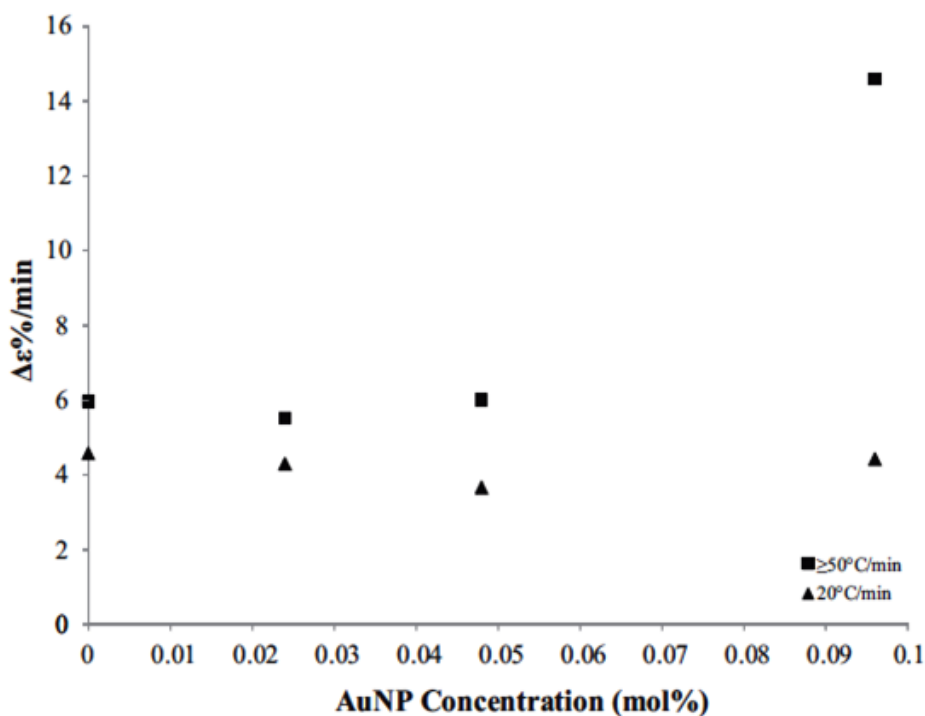


Fig.14: LCE samples contain 0.096mole% AuNPs improve the actuation speed more than 100% compared with sample do not contain AuNPs [26].

Future Work and Conclusions:

This was a short review to summarize the current knowledge of important properties of LCE since light energy can be converted into mechanical power directly using deformations of photodeformable polymers. However, future research must to be carried out in order to control the shape changes of LCE materials electrically. To this end, one strategy is to swell LCEs using low molecular-weight liquid-crystal materials, which raise the liquid crystalline cooperativity beyond the normal LCE mesh size. When an electrical current pass through such a sample, its temperature increases, initiating a shape change.

LCEs are promising materials for the construction of artificial muscles driven by light. Despite this, the photomechanical characteristics of such materials require further development, particularly the mesogenic alignment, the coupling of LC order with polymer networks, and the change in order induced by the light. The issues of fatigue resistance and biocompatibility of these materials must also be addressed. LCEs are not suitable for steady-state applications since thermal activations can continue, consuming substantial power even in a no-work condition. Finally, it is expected that in the future, LCEs will be used in new generations of silicon photovoltaics used to convert solar energy to electricity.

This work first described the thermal properties of LCEs. It was shown that the thermal properties of LCEs can be affected by the overall flexibility of the polymer backbone, the nature of the mesogenic monomer, and both the type and degree of crosslinking (which has an impact on the glass transition temperature). Following this, the mechanical properties of LCEs were reviewed. LCE polydomains are usually opaque, but the application of a large enough uniaxial extension induces a change to a transparent, monodomain material state. The response of soft LCE materials to light, electric fields, and thermal energy was also studied, and a mechanical response to such inputs was observed. It is found that LCE materials have the ability to change their shapes by changing direction of LC director when they are exposed to light, electric field and thermal energy. Further, it was found that in swollen samples, the threshold field was reduced by a factor of 200 when compared to unswollen samples. Additionally, this paper shows that the thermo-mechanical property of LCEs can be highly affected by addition of nanoparticles to the sample. These data provide evidence that LCEs are attractive materials for fabrication of soft actuators and artificial muscle, allowing large magnitude,

high-speed deformation and either two-dimensional (contraction) or three-dimensional (bending) transformation.

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