Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713926090

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To cite this Article Hikmet, R. A. M. and Poels, H. L. P.(2000) 'An investigation of patterning anisotropic gels for switchable recordings', Liquid Crystals, 27: 1, 17 — 25
To link to this Article: DOI: 10.1080/026782900203164
URL: http://dx.doi.org/10.1080/026782900203164

LIQUID CRYSTALS, 2000, VOL. 27, NO. 1, 17–25

An investigation of patterning anisotropic gels for switchable recordings

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(*Received 12 May 1999; accepted 19 June 1999*)

A new kind of anisotropic gel has been produced by photopolymerization of liquid crystal (LC) mixtures composed of monoacrylates, diacrylates and non-reactive conventional LC molecules. After polymerization, lightly cross-linked anisotropic polymer networks swollen by the non-reactive molecules are produced. These systems showed a gel-point associated with the onset of the formation of a three dimensional network. Above this point, a rapid increase in the threshold voltage was observed. Various factors, such as the photoinitiator concentration and the monoacrylate and diacrylate concentrations, as well as the intensity of the light used for the polymerization, have a strong influence on the gel-point. The switching kinetics of the materials are also strongly influenced by the aforementioned parameters. Two relaxation processes were found to occur within the gels above the gel-point. Using dielectric spectroscopy, this behaviour was found to be associated with an inhomogeneous phase structure within the gels. Patterned irradiation was used to produce regions with different threshold voltages for switching. In this way, patterns were created in the gels, which become visible, by the application of an electric field. It was demonstrated that this technique can be used to produce switchable gratings, Fresnel lenses, and photographs.

1. Introduction

Anisotropic gel are obtained by *in situ* photopolymerization of reactive molecules in the presence of non-reactive LC molecules. Gels obtained by polymerization in various LC phases have been described in previous articles [1–4]. These gels were obtained using bifunctional reactive molecules which formed cross-linked networks. It was found that LC molecules in these gels which were not chemically attached to the network could be switched under the influence of electric fields, while the orientation of the network remained unaltered due to its rigid nature. The use of these gels in very fast electrically-induced scattering with no haze in the field-off state has been demonstrated [1]. Recently, a new type of anisotropic gel composed of a cross-linked side group liquid crystal polymer and a non-reactive LC has been described [5]. In such gels, the cross-linked network provides the system with a memory function and facilitates reversal to the initial orientation state after switching. In the case of cholesteric gels for example, fast switching between a defect-free colourful planar state and a transparent homeotropic state proved possible.

It has also been shown that in gels the network density determines the threshold of switching. We have now investigated the possibility of using such gels as switchable recording media. It was expected that illumination with a modulated light intensity would lead to a network representing the light intensity distribution used for the gel formation as a result of diffusion of polymerizable groups within the monomeric mixture. This distribution can be made visible by applying an electric field across such a gel in the form of a refractive index distribution. Switchable gratings and lenses based on a liquid crystal cell with patterned transparent indium tin oxide (ITO) electrodes have already been demonstrated [5]. Here we would like to demonstrate the possibility of producing such switchable devices by patterning the gels instead of the electrode. Properties of such gels and the parameters to produce such devices will also be described.

2. Experimental

The structures of the monoacrylate 4-(6-acryloyloxyhexyloxy)-4′-cyanobiphenyl (CB6) and the diacrylate C6M used for cross-linking have been described in previous publications [7]. The conventional LC E7 used in the experiments was purchased from Merck Ltd (Poole). The pre-gel mixtures were provided with the photoinitiator Irgacure 651 in order to facilitate photopolymerization. Mixtures were oriented in cells provided with uniaxially buffed polyimide layers and conducting transparent orientation layers. The cell gap was kept at

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Liquid Crystals ISSN 0267-8292 print/ISSN 1366-5855 online © 2000 Taylor & Francis Ltd
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6 μm. Polymerization was initiated using a Philips PL lamp with emission at 350 nm. Use was also made of a Spectra Physics 2020 Krypton laser in UV mode in experiments in which grating and Fresnel structures were produced by illuminating appropriate masks. The wavelengths of this laser were 354.8 nm (80% power) and 359 nm (20%). Dielectric measurements were carried out using a Hewlett-Packard 4194A Impedance/Gain-Phase Analyser. An optical polarizing microscope provided with a stable light source and a photomultiplier were used for the electro-optic measurements. Birefringence measurements were carried out using a rotary compensator.

3. Results and discussion

3.1. *Electro-optical properties of the mixtures*

Gels were produced by polymerization in the uniaxially oriented state. In order to investigate the switching behaviour of the gels, we measured their effective birefringence as a function of the voltage by determining the monochromatic light ($\lambda = 546$ nm) intensity (I) transmitted through crossed polarizers with the uniaxially oriented gels placed between them at an angle of 45° during the application of an electric field. Figure 1 shows an example of the curves obtained. Equation (1) shown below was used to plot the measured curves as effective birefringence (Δn_{eff}) versus voltage.

$$I = I_{\max} \sin^2(\pi d \Delta n_{\mathrm{eff}}/\lambda). \tag{1}$$

Figure 2 shows the effective birefringence as a function of the voltage for various gels containing various amounts of network molecules. In each curve, a threshold voltage can be identified corresponding to the point at which the effective birefringence began to decrease. It can also be seen that the threshold voltage remained constant up to a certain diacrylate (C6M) concentration, above which the threshold voltage rapidly increased. The threshold voltage (V_c) of a uniaxially oriented system is given by equation (2).

$$V_c = \pi(K_1/\varepsilon_o \Delta\varepsilon)^{0.5} \tag{2}$$

where K_1 is the splay elastic constant, ε_o is the permittivity of free space and $\Delta\varepsilon$ is the dielectric anisotropy of the material. The fact that the threshold voltage shows an increase above a critical concentration indicates that the elastic constant shows an increase above this concentration corresponding to the gel-point of the system. At this concentration a three dimensional network is created by the side group polymers formed by the monoacrylate molecules cross-linked by the diacrylate molecules. Here it is important to note that the other factor $\Delta\varepsilon$ which can influence the threshold voltage was found to show almost no change with increasing C6M concentration.

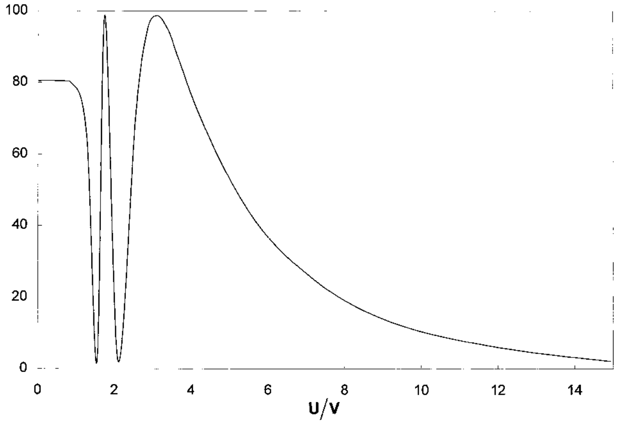


Figure 1. Transmission as a function of the voltage for 10 wt % CB6; 0.63 wt % C6M.

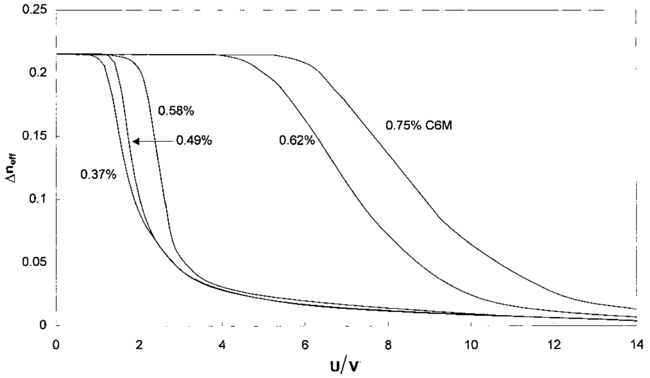


Figure 2. Effective birefringence as a function of the voltage for gels containing 10 wt % CB6 and various wt % of C6M.

The gel formation is schematically represented in figure 3. In this schematic representation it can be seen that the factors influencing the gel-point are the cross-link density and the kinetic chain length of the linear molecule. The cross-link density is determined by the concentration of the diacrylate molecules within the system, as shown in figure 3. The kinetic chain length is on the other hand determined by various parameters. One of the parameters is the concentration of the monoacrylate.

In figure 4 the threshold voltage V_c is plotted as a function of C6M concentration for gels obtained by polymerizing various amounts of the monoacrylate. The initiator concentration in the mixtures was 0.5% and the intensity of the UV light was 1 mW cm^{-2}. As expected, it can be seen in this figure that the amount of diacrylate required to reach the gel-point decreases with increasing monoacrylate concentration. Furthermore, for gels with high polymer concentrations the increase in the V_c above

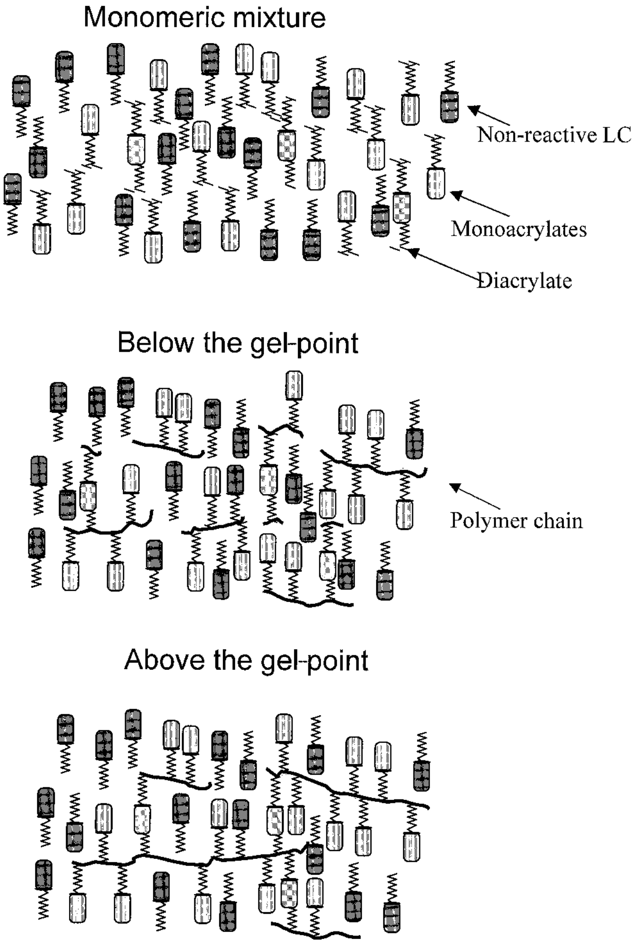


Figure 3. Schematic representation of gel formation.

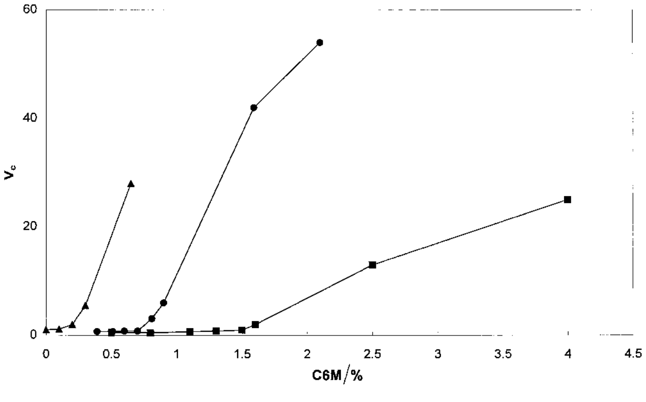


Figure 4. Threshold voltage as a function of wt % C6M concentration for gels containing various wt % amounts of CB6 at a constant intensity of 1 mW cm^{-2} and 0.5 wt % photoinitiator. ■ = 5 wt % C6M, ● = 10 wt % C6M, ▲ = 20 wt % C6M.

the gel-point is much faster than in gels with lower monoacrylate contents. This behaviour can also be associated with a higher concentration of pendant groups in the systems with lower concentrations of monoacrylate.

The other parameters determining the kinetic chain length are the intensity of the UV light and the concentration of the photoinitiator. These parameters determine the number of initiation sites and hence the chain length. We investigated each of these parameters separately. The results are shown in figures 5 and 6. In figure 5 the threshold voltage is plotted as a function of the initiator concentration for various gels containing different amounts of C6M cross-linker and 10 wt % CB6. It can be seen that the threshold voltage increases with a decreasing network concentration in the case of all the gels, as expected. In all these curves the threshold voltage remains almost constant down to a certain initiator concentration and shows a continuous increase below it. Furthermore, when mixtures containing various amounts of C6M are compared, it can be seen that the gel-point shifts to higher initiator concentrations with

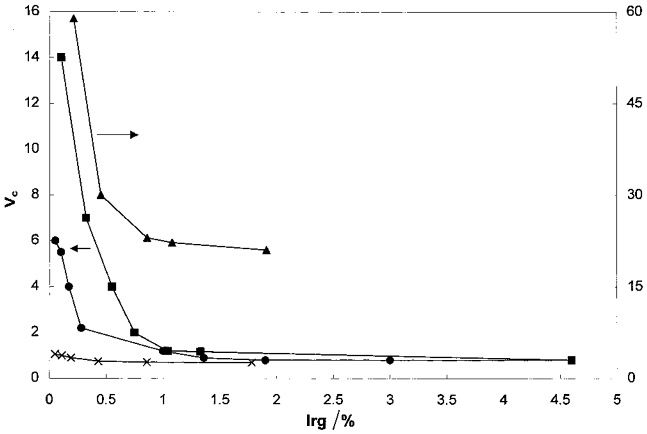


Figure 5. Threshold voltage as a function of the photoinitiator (Irgacure) concentration (wt %) for gels containing 10 wt % CB6 and various amounts of C6M produced at 1 mW cm^{-2}. x = 0.4 wt % C6M, ● = 0.63 wt % C6M, ■ = 0.8 wt % C6M, ▲ = 1.5 wt % C6M.

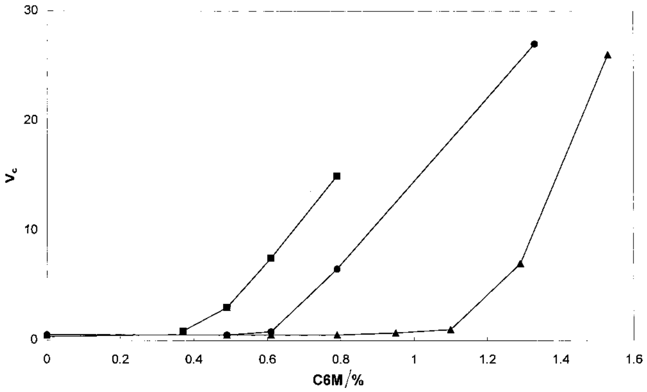


Figure 6. Threshold voltage as a function of the UV intensity for gels containing 10 wt % CB6, 0.5 wt % Irg, and various amounts of C6M. ■ = 0.1 mW cm^{-2}, ● = 1 mW cm^{-2}, ▲ = 8 mW cm^{-2}.

increasing C6M concentration. This indicates that when the cross-linker (C6M) concentration decreases the kinetic chain length has to be increased in order to reach the gel-point.

The effect of light intensity can be seen in figure 6, which shows the threshold voltage as a function of C6M concentration for gels obtained by polymerization under various UV intensities. The results shown here can be interpreted in the same manner as those presented in figure 5. Here, the intensity of the UV light is the determining factor for the kinetic chain length instead of the initiator concentration. In figure 6 the gel-point again appears at higher light intensities and higher concentrations of the network, indicating that the kinetic chain length decreases with increasing UV intensity.

Within the gels, not only the threshold voltage, but also the dynamics of switching are influenced by the presence of the network. We used the method described by Wu and Wu [8] to study the switching behaviour within the gels. For this purpose, a small voltage was applied across the gel to induce an average tilt of 15° of the LC molecules within the cell. This resulted in a decrease of δ_o in the phase difference within the cell. After removing the voltage and short-circuiting the cell the retardation change is followed as a function of time, $\delta(t)$. According to Wu and Wu [8], the retardation changes from $\delta(t) = \delta_o$ at time $t = 0$ to $\delta(t) = 0$ at $t = \infty$ and can be represented as

$$\delta(t) = \delta_o \exp(-2t/\tau) \qquad (3)$$

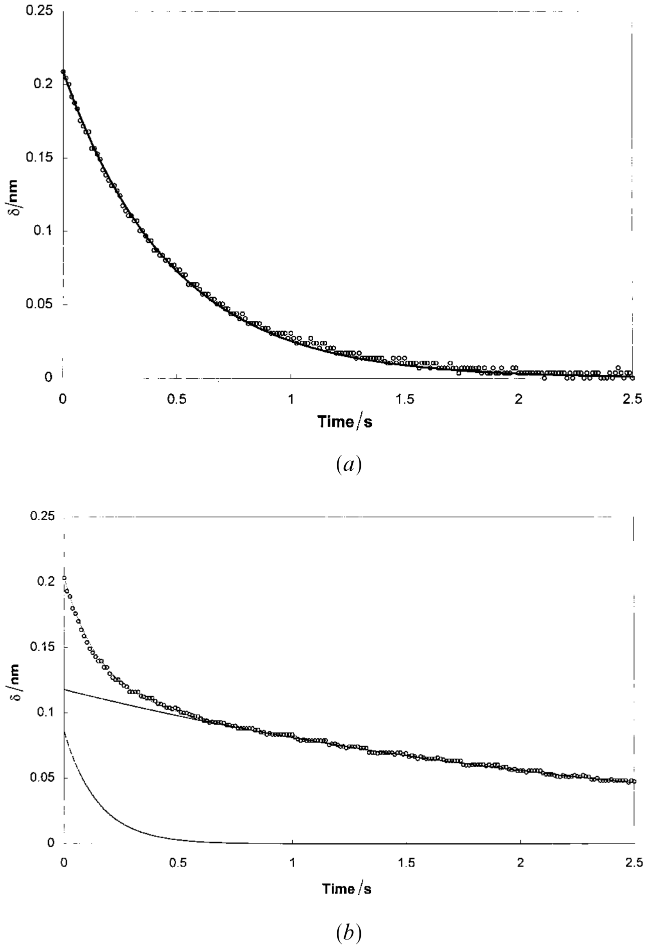
where the decay constant τ is related to the other physical constants of the material as shown below

$$\tau = \gamma d^2/\pi^2 K_1 \qquad (4)$$

where d is the cell thickness and γ is the bulk rotational viscosity.

In figures 7(*a*) and 7(*b*), $\delta(t)$ is plotted as a function of time for gels containing different amounts of network. It can be seen that in the system containing 0.4 wt % C6M (which lies below the gel-point) the retardation decay followed a single exponential decay function, represented by equation (3). In the case of the gel containing 0.8 wt % C6M, a fast change in retardation was however followed by a slow decay. This behaviour can be represented by the sum of two exponential decay functions with two different decay constants. The existence of two decay constants in the gel indicates that there are two different relaxation processes, described by two decay constants, τ_s and τ_f, corresponding to the slow and fast switching mechanisms, respectively.

Before elaborating on the causes of this behaviour, we will take a closer look at the behaviour observed in various gels. Figure 8 shows the influence of the cross-link density on the decay constants for gels containing



(*a*)

(*b*)

Figure 7. Retardation as a function of time for gels containing 10 wt % CB6 and (*a*) 0.4 wt % C6M, (*b*) 0.81 wt % C6M. Solid lines represent the theoretical fit.

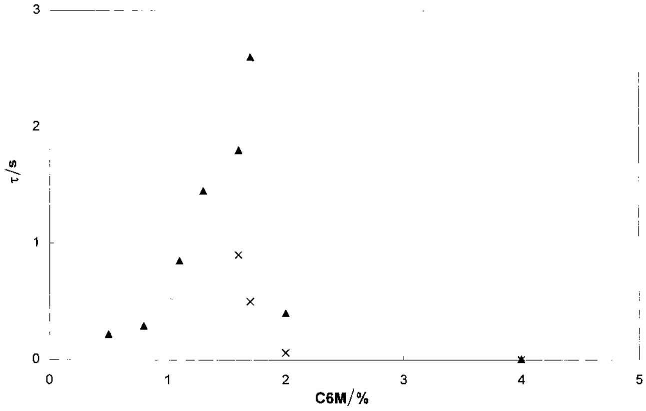


Figure 8. Decay constants τ_f (crosses) and τ_s (triangles) as functions of C6M concentration (wt %) for a system containing 5 wt % CB6 and 0.5 wt % initiator irradiated at 1 mW cm^{-2}.

5 wt % CB6. It can be seen that the decay time increases with increasing network concentration until the cross-link concentration for gel-point is reached. Above this point, the two decay constants become indentifiable and both decrease with an increasing cross-link density (C6M concentration). The increase observed in the decay constant with an increasing C6M concentration can be associated with an increase in the viscosity of the system as a result of the formation of an increasingly branched, high molecular mass polymer. The presence of the acrylate backbone in the system greatly slows down the reorientation of the mesogenic side groups, resulting in an increased bulk rotational viscosity.

Above the gel-point, two factors contribute to the decrease in the decay constant. These factors are: (i) an increase in the elastic constant due to the formation of the network, (ii) a decrease in the internal viscosity as the network to a great extent prevents the reorientation of the polymer backbone. The presence of the two decay constants above the gel-point can be explained in the same way. Above the gel-point, the gel consists of a network swollen by the low molar mass LC molecules. In regions within the gels in which the polymer chains are cross-linked, the main polymer chain motion is restrained, so the reorientation of the mesogenic groups can take place without a high degree of reorientation of the main chain. These regions hence have a lower rotational viscosity than regions in which the polymer chains are attached to the network as pendant groups. Motion of such pendant polymer groups causes an increase in the rotational viscosity. We tried to estimate the rotational viscosity of the gels using the equation below.

$$\gamma = (\tau \varepsilon_o \Delta\varepsilon V_c^2)/d^2. \qquad (5)$$

For this purpose we measured the dielectric constants of the gels where the molecules were oriented in the direction parallel ($\varepsilon_\parallel$) or perpendicular ($\varepsilon_\perp$) to the electric field. We then used these values to calculate the dielectric anisotropy $\Delta\varepsilon$. In figure 9 the dependence of the dielectric constants of various gels is shown as a function of the temperature. It can be seen that an increase in the CB6 concentration caused a slight increase in the dielectric anisotropy. However, the addition of up to 2 wt % C6M to the gels did not result in any significant change in the dielectric anisotropy. Using the measured values of threshold voltages and dielectric anisotropy, the rotational viscosity γ was calculated by inserting the decay constant obtained from figure 8. The results are shown in figure 10. It can be clearly seen that the rotational viscosity increased with increasing C6M concentration up to the gel-point. Above the gel-point two rotational viscosities were calculated according to equation (5) using the values of τ_s and τ_f obtained from figure 8. The validity of the equation in relation to network formation is not quite clear, as the switching behaviour is then determined by the anisotropic network instead of by the cell surfaces. We nevertheless calculated the rotational viscosity above the gel-point. It can be seen that the rotational viscosity decreases above the gel-point, but does not reach the level observed for low concentration of C6M.

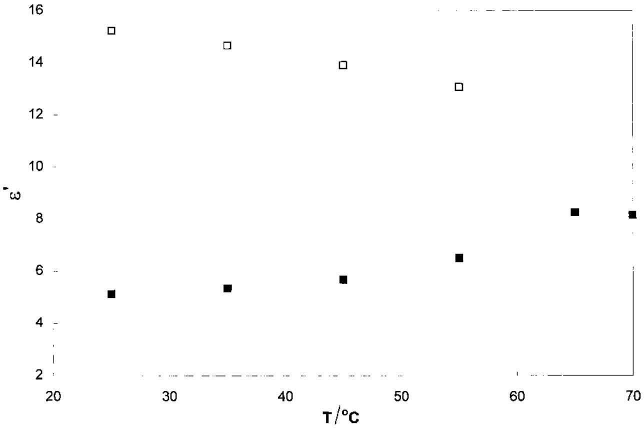


Figure 9. Dielectric constants as a function of temperature measured for 10 wt % CB6 and 0.6 wt % C6M. ■ = $\varepsilon_\perp$, □ = $\varepsilon_\parallel$.

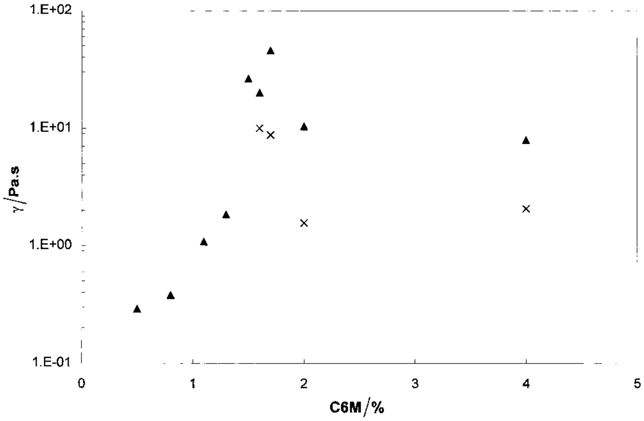


Figure 10. Rotational viscosities γ_f (crosses) and γ_s (triangles) as functions of C6M concentration (wt %) for a system containing 5 wt % CB6 and 0.5 wt % initiator irradiated at 1 mW cm^{-2}.

Having shown the importance of the cross-link density on the switching kinetics, we went on to investigate the effect of the kinetic chain length by varying the amounts of initiator used during polymerization of systems containing 10 wt % CB6 and various amounts of C6M. Figures 11 (*a*) and 11 (*b*) show τ_f and τ_s as functions of the initiator concentration. The calculated rotational viscosities of the systems are shown in figures 12 (*a*) and 12 (*b*). In these figures, zero initiator concentration represents the behaviour of the monomeric mixture. The

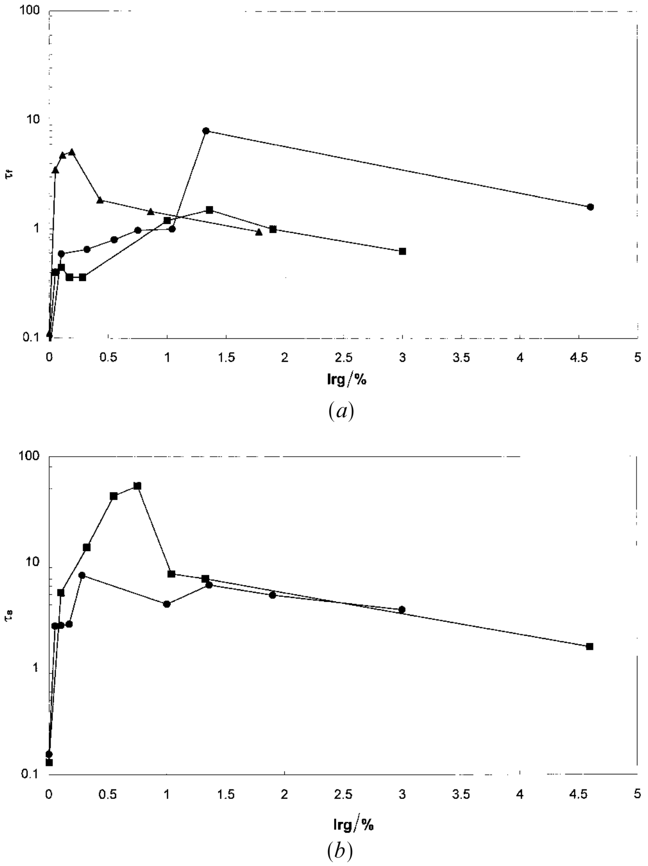


Figure 11. Decay constant (*a*) τ_f and (*b*) τ_s as functions of initiator (Irgacure) concentration for systems containing 10 wt % CB6 and various amounts of C6M irradiated with $1\,\mathrm{mW\,cm^{-2}}$. ■ = 0.4 wt % C6M, ● = 0.63 wt % C6M, ▲ = 0.8 wt % C6M.

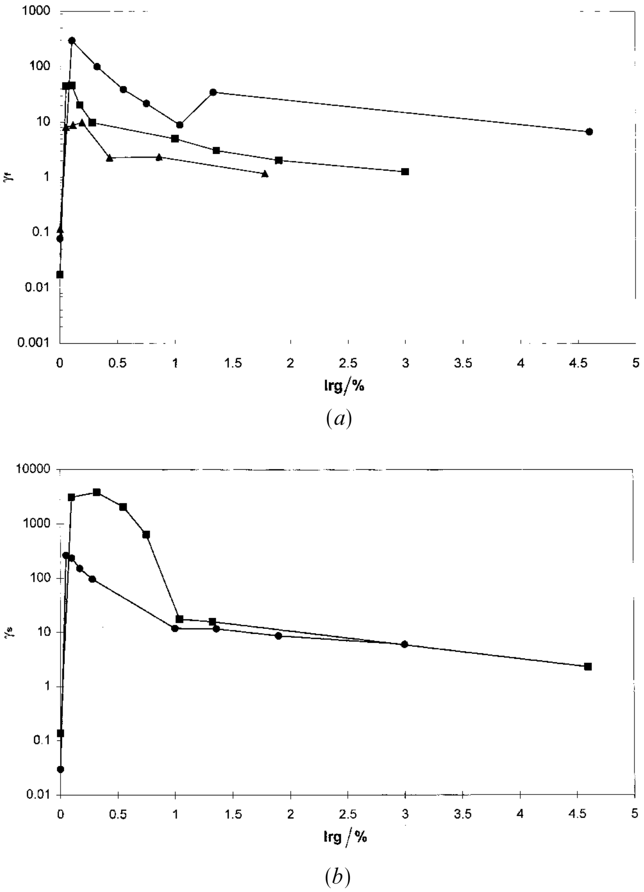


Figure 12. Rotational viscosities (*a*) γ_f and (*b*) γ_s as functions of initiator (Irgacure) concentration for systems containing 10 wt % CB6 and various amounts of C6M irradiated at $1\,\mathrm{mW\,cm^{-2}}$. ■ = 0.4 wt % C6M, ● = 0.63 wt % C6M, ▲ = 0.8 wt % C6M.

increase observed relative to this point therefore merely indicates the effect of polymer formation, resulting in an increase in the internal viscosity.

In the system, containing 0.4 wt % C6M a single decay constant τ was found to be effective, as this system lies below the gel-point for all initiator concentrations. In this system, the viscosity shows a steady decrease with increasing initiator concentration above the zero initiator concentration, indicating a lower viscosity as a result of reduced kinetic chain length.

In figures 11 and 12 it can be seen that the mixtures containing 0.6 wt % and 0.8 wt % C6M showed a rather different behaviour than the gel containing 0.4 wt % C6M. Up to initiator concentrations of 1 wt % and 0.5 wt %, corresponding to the gel-points of the mixtures containing 0.8 wt % and 0.6 wt % C6M, respectively, τ_f remained almost constant, whereas it increased above these points. On the other hand, τ_s increased with increasing initiator concentration up to the respective gel-points of the gels and then decreased above these points to be almost the same at higher initiator concentrations. The calculated rotational viscosity plots show the same trends and the viscosity remains almost constant up to the gel-point before showing a steady decrease with increasing initiator concentration. The observations reported here are in accordance with the hypothesis that the kinetic chain length and the nature of the network determine the switching kinetics of these systems.

We used dielectric spectroscopy to take a closer look at the switching behaviour of the gels. The relaxation times associated with the motion of the molecules around their short axes (δ-process) were measured in a state in which the molecules were aligned parallel to the applied field. Figure 13 shows the dielectric loss (ε'') measured at various temperatures in a gel containing 20 wt % CB6 and 0.4 wt % C6M. It can be seen that with decreasing temperature the relaxation peak shifts to lower temperatures and decreases in height as it becomes broader. When the dielectric constant was plotted as a function

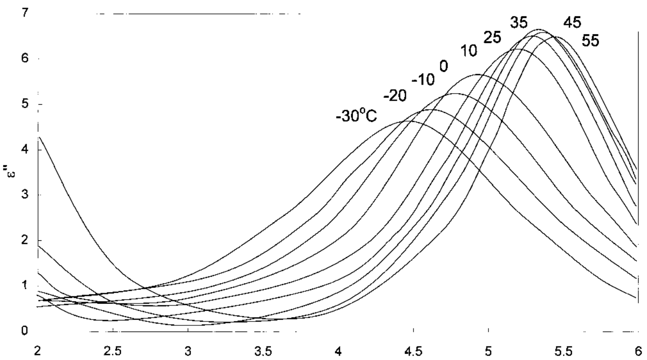


Figure 13. Dielectric loss as (ε'') as a function of frequency for a gel containing 20 wt % CB6 and 0.4 wt % C6M; and as a function of frequency measured at various temperatures.

of dielectric loss (Cole–Cole plot) an almost perfect semi-circle was obtained for pure E7, indicating that a single mean relaxation time was effective within this system. In the case of the gels, the Cole–Cole plots however were depressed semi-circles, indicating that within the gels a distribution of mean relaxation times is operational.

By taking the frequency corresponding to the maximum in the dielectric loss as the relaxation frequency, we produced the Arrhenius plot shown in figure 14. This plot clearly shows non-linear behaviour. In the case of a homogeneous liquid crystal far from the glass transition temperature, such an Arrhenius plot would be expected to be a straight line, representing the free volume effect on the mobility within the system. The non-linear behaviour observed in this figure indicates the presence of a phase-separated system, which becomes more homogeneous with increasing temperature. In a system consisting of two phases, mixing at higher temperatures can lead to a change in the composition of the phases. Therefore, a change in temperature can cause the mean relaxation time to change too, as a result of the change in the mobility, but also the change in the composition of the phases.

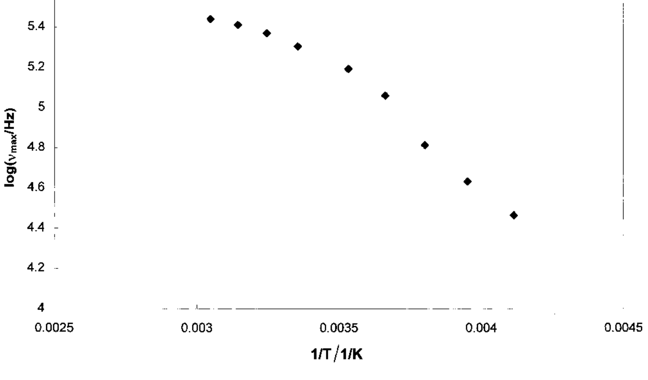


Figure 14. Arrhenius plot of dielectric relaxation.

3.2. *Patterned irradiation of gels*

Using some of the mixtures described above we tried to produce patterned gels. For this purpose, cells with transparent electrodes were used and the mixture was irradiated with UV in two stages. In the first stage the mixture was irradiated through a mask containing ruled gratings. In the second state the mask was removed and the whole cell was exposed to flood radiation. Two effects play a role in this process: (1) in the first step, the gel becomes polymerized in the irradiated areas, causing the diffusion of reactive groups to these areas; (2) in the second step, all the molecules in the system are polymerized. As the first step is limited by diffusion, the reactive mixture has to be irradiated at a low intensity in the first step and a high intensity in the second step when large structures have to be produced. When a gel containing 0.5 wt % photoinitiator, 10 wt % CB6 and 0.6 wt % C6M was irradiated at a UV intensity of 0.0125 mW cm^{-2}, line widths of up to 1250 µm were obtained upon exposure through a mask. Figure 15 shows an optical photograph of a mask together with photos of the gel at various stages of applied voltage observed between crossed polarizers. The transparent areas of the mask, where polymerization took place during the first stage of UV radiation, led to the formation of a network within the gel with a high threshold voltage. This explains why, when an electric field was applied across the gel, these areas did not switch, and only the areas which were irradiated during the second state of UV radiation started to switch, figure 15(*c*). As the voltage increased, these areas showed different birefringence colours, figure 15(*d*), before becoming dark, figure 15(*e*). When the voltage was increased even further, the areas with a higher threshold voltage irradiated in the first state started to switch as can be seen in figure 15(*f*). The diffraction efficiency of this switchable grating was studied by measuring the intensity of a laser beam (540 nm). In figure 16 the intensities of the zero and first order spots are shown as functions of the applied voltage. It can be seen that the intensity plots obtained for the peaks are out of phase and passed through maxima and minima as the voltage increased. The high degree of diffraction efficiency indicates that the line profiles were retained during the switching.

Having shown that switchable gratings can be produced we also tried to produce more complicated patterns such as binary-phase Fresnel lenses [9]. For this purpose, a mask was produced containing the binary zones in the form of concentric rings of dark and

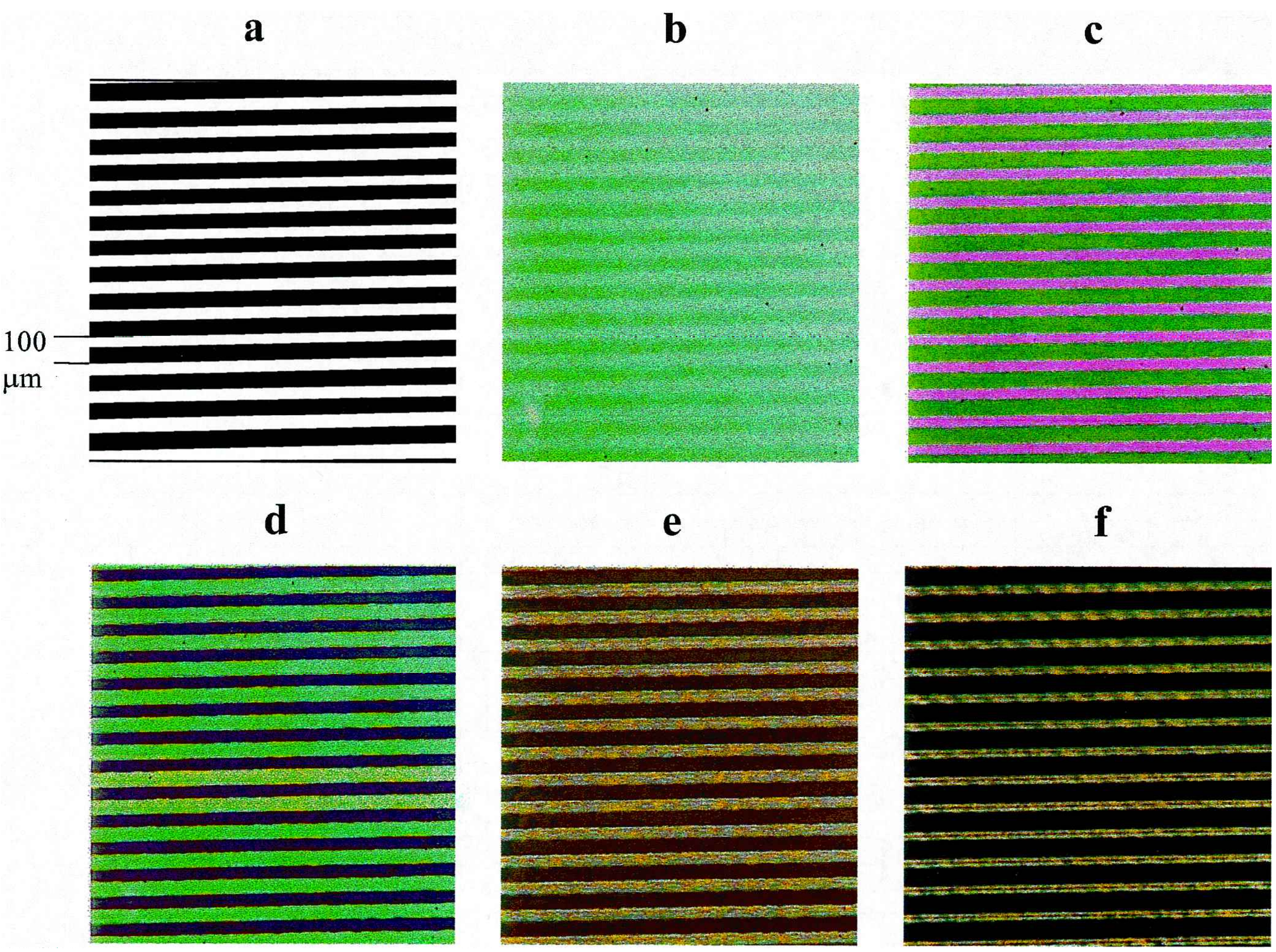


Figure 15. Photographs of the grating (*a*) and the gel at various applied voltages: (*b*) 0 V, (*c*) 1.2 V, (*d*) 12 V, (*e*) 32 V, (*f*) 70 V.

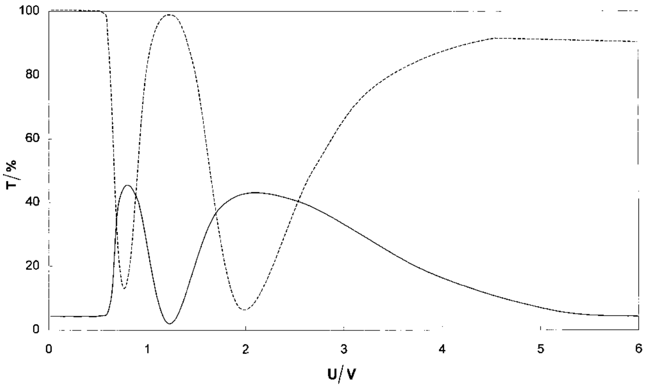


Figure 16. Intensity of the zero and first order diffraction peaks during the application of an electric field across the gel. ---- Zero order, __ first order.

transparent regions. In such a mask the radius of the nth zone R_n is given by $R_n = nR_1^2$, where R_1 is the radius of the innermost zone. The relaxation between the focal length f of the lens and R_1 is $f = R_1^2/\lambda$, where λ is the wavelength of the light used. The gel was structured by irradiating it through the mask. The cell obtained was used to focus a broad beam of laser light to a point corresponding to the focal point of the lens. The intensity of the focused beam increased with increasing voltage, before decreasing at higher voltages. As explained above this is because maximum diffraction efficiency is obtained when the phase difference between the two zones is $\phi = m2\pi\Delta nd/\lambda$, where d is the gel thickness and m is an integer. Thus, a switchable zone plate lens with a focal length of 80 cm was obtained.

We also explored the possiblity of introducing more complicated structures into such gels. We used a photonegative and irradiated the cell through it. Figure 17 shows the photograph of the cell placed between crossed polarizers with and without applied voltage. It can be seen that the image used for patterning the gel became visible upon application of a voltage, showing that a gel can be fed with complicated structures.

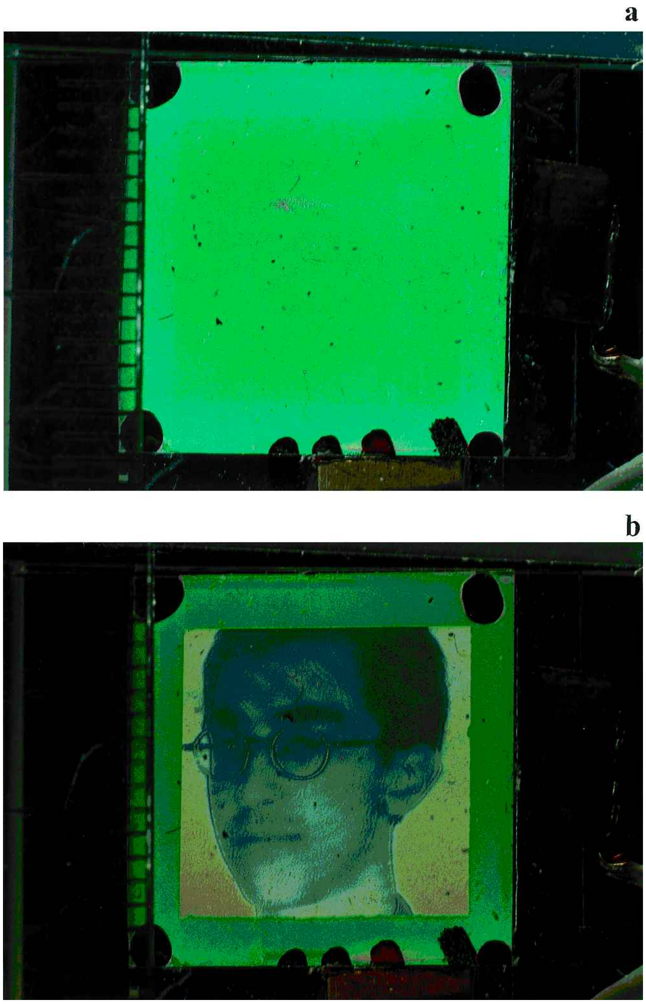


Figure 17. Photographs of the recordings made in the gel (*a*) before and (*b*) after the application of a voltage.

4. Conclusions

The *in situ* production of a new type of macroscopically oriented anisotropic gel consisting of a lightly cross-linked side group polymer swollen by non-reactive conventional LC molecules has been demonstrated. It has been found that the gels can be switched under the influence of an electric field and that the molecules revert to their initial state of orientation when the electric field is removed. It was found that the threshold voltage for the switching starts to show a sharp increase at the gel-point at which a three dimensional network is formed. The gel-point in these gels was found to be influenced by variables such as the intensity of the UV light, and the initiator and diacrylate concentration. At a given intensity, with increasing initiator concentration, higher amounts of diacrylate are needed in order to reach the gel-point. Accordingly, at a given initiator concentration, higher diacrylate concentrations are needed to reach the gel-point with increasing light intensity. This behaviour has been explained in terms of a decrease in the kinetic chain length with increasing light intensity and initiator concentration. Above the gel-point switching behaviour of the gels also changes; it then occurs at two rates, whereas it switches at a single rate below the gel-point. The two rates observed for the gels are attributed to the inhomogeneous nature of the network confirmed by the dielectric relaxation measurements.

The sensitivity of the gel-point to the intensity of the UV light and the ability of the reactive components to diffuse during gel formation makes this system suitable for use as a recording medium. Patterned irradiation of the mixture with UV light led to the formation of gels characterized by regions with different threshold voltages. In this way recordings can be made in gels and made visible upon application of an electric field in the form of regions with different birefringence.

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