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## Liquid Crystals

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#### Liquid crystals for active matrix displays

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Active matrix liquid crystal displays belong to one of the fastest growing fields in display research and development. Several pocket TVs using this technology are already available commercially. High quality displays for TV and instrumentation require an understanding and optimization of liquid crystal material parameters. We have, therefore, investigated the influence of the elastic, dielectric and optic properties on the electrical resistance of the liquid crystal as well as on the response times and the viewing angle dependence of active matrix displays. Based on correlations derived from our investigations, we have developed new liquid crystals which have suitable properties and a very high electrical resistivity.

#### 1. Introduction

Active matrix addressing refers to any array of non-linear elements used to switch individual picture elements, or pixels, in a liquid crystal display (LCD). Although amorphous-Si TFTs now dominate the market, other promising technologies are poly-Si TFTs and diode arrays (MIM or back-to-back a-Si diodes). Liquid crystal TV displays with active matrix addressing have attained properties which make them one of the most important candidates for large area, flat TV screens. A considerable disadvantage at the moment is the high price of the active matrix, which is due mainly to the low production yield.

Today practically all liquid crystal TV displays are working as a twisted nematic cell (TNC). If we look at such a cell closely, we can recognize that this type of display has some fundamental problems in addition to the high price. These are especially the viewing angle dependence of the contrast [1-3], and the response times and insufficient specific resistances of the liquid crystal material [4, 5]. We have tried to establish the most important liquid crystal parameters which control these problems. In doing this we have compared experimental results with results obtained by computer simulations. Based on this work, new liquid crystal mixtures were developed which help to improve these problems. However, as an additional result of our work, it has been clearly demonstrated that a TNC in the first minimum, according to Gooch and Tarry [6, 7], is the most favourable for a TV screen [8].

#### 2. Viewing angle dependence $\Delta C$

As a display principle, which has an adequate viewing angle dependence, the guest-host display is discussed. However, problems with the relatively low specific resistivity of such liquid crystal mixtures and limited light stability of the dyes have still not been solved. Recently, a double layer supertwisted nematic (STN) display was proposed. However, in this case, despite the excellent viewing angle dependence, decisive disadvantages are the limited response times and probably also the steep contrast curves making an accurate control of grey scale levels difficult.

In consequence, twisted nematic displays are still the favourite for active matrix displays. Consequently, all electro-optic measurements were performed in TNCs with a twist angle of 90°. The tilt angle was of the order of 1° to 2°. There are four possibilities for the positioning of the polarizers:

- (a) crossed polarizers, rear and front polarizer parallel to the liquid crystal director,
- (b) crossed polarizers, rear and front polarizers perpendicular to the liquid crystal director,
- (c) parallel polarizers, near polarizer parallel to the liquid crystal director,
- (d) parallel polarizers, rear polarizer perpendicular to the liquid crystal director.

Configurations (a) and (b) lead to a positive contrast, (c) and (d) result in a negative contrast. According to our definition of the viewing angle dependence  $\Delta C$ , given later, cells of type (b) exhibit a better viewing angle dependence than cells of type (a), cells (c) and (d) are equal and rank between (a) and (b) (these experimental results are in good agreement with computer simulations). We decided, therefore, to use configuration (b) (in transmission) for all our measurements.

We have performed two series of measurements. In the first, six liquid crystal mixtures, which differ largely in their optic, dielectric and elastic properties (see table 1), have been investigated over a range of optical retardations  $d\Delta n$  from about 0.4 to 2  $\mu$ m (here *d* is the cell gap and  $\Delta n$  the optical birefringence of the liquid crystal). In the second series, a larger number of about 20 liquid crystal mixtures were investigated at constant  $d\Delta n$  of exactly 0.5  $\mu$ m (the maximum deviation was 0.01  $\mu$ m). These liquid crystal mixtures differ in their properties in the same way as the mixtures from the first series.

Table 1.	Properties	of	the	liquid	crystal	mixtures	used	in	the	first	and	second	series	of
measurement.														

Clearing point, $T_{\rm NI}/^{\circ}\rm C$	80-100
Viscosity, $\nu/\text{mm}^2 \text{s}^{-1}$	18-21
Optical anisotropy, $\Delta n$	+0.07 to $+0.16$
Dielectric anisotropy, $\Delta \varepsilon$	+2  to  +11
Elastic constants	
$K_{11}/10^{-12}\mathrm{N}$	10.2-12.6
$K_{22}/10^{-12}\mathrm{N}$	5.7-10.5
$K_{33}/10^{-12}\mathrm{N}$	12.9-24.0

The definition of the viewing angle dependence  $\Delta C$  used in this work is given in figure 1. It has been found to be useful to characterize the viewing angle dependence of a TNC used as a TV screen. It is assumed that a certain pixel shows a relative contrast of 50 per cent at a viewing angle of 10°. If the viewing angle is changed to 0°, the relative contrast decreases to a value of C(0), whereas the relative contrast increases to C(30) when the viewing angle is changed to 30°. The difference between C(0) and C(30) is the measure which we use for the viewing angle dependence, called  $\Delta C$ . This value should be as small as possible. Figure 2 shows the viewing angle dependence  $\Delta C$  of several mixtures versus  $d\Delta n$ . It can be seen that  $\Delta C$  improves significantly with decreasing  $d\Delta n$ . This is in good agreement with computer simulations, performed by Baur *et al.* [9]. For all mixtures, investigated here  $\Delta C$  is about 40 per cent for a  $d\Delta n$  of  $0.5 \,\mu$ m (first minimum according to Gooch



Figure 1. Definition of the viewing angle dependence,  $\Delta C$ .



Figure 2. Viewing angle dependence,  $\Delta C$ , as a function of  $d\Delta n$  (at 20°C). The different symbols represent six different liquid crystal mixtures (series 1, see text).

and Tarry [6, 7]). We can also recognize that there are no significant differences between the different mixtures.

We have tried to establish correlations between the important liquid crystal parameters and the viewing angle dependence,  $\Delta C$ . However, for all mixtures investigated here, including mixtures from both series, we could not find, for example, a correlation between the elastic constants and  $\Delta C$ . Mixtures with the same dielectric and optic properties, but with significantly different ratios of  $K_{33}/K_{11}$ , do not show different viewing angle dependences. For example,  $K_{33}/K_{11}$  values of 1.5 and 0.5 (the other properties being very similar) lead to the same  $\Delta C$ .

If we compare the mixtures from series one and two with regard to  $\Delta \varepsilon / \varepsilon_{\perp}$ , there is no example where we find the same  $\Delta \varepsilon$  at different  $\Delta \varepsilon / \varepsilon_{\perp}$  values. All mixtures investigated in this work follow the rather strict correlation between  $\Delta \varepsilon / \varepsilon_{\perp}$  and  $\Delta \varepsilon$  as illustrated in figure 3. The new stable 2,3-diffuoro-1,4-phenylene derivatives described by Reiffenrath *et al.* [10] now enable us to increase  $\varepsilon_{\perp}$  without increasing the viscosity, while the specific resistance is kept at a high level. This leads to a smaller  $\Delta \varepsilon / \varepsilon_{\perp}$  at the same  $\Delta \varepsilon$ . However, this does not result in an improved viewing angle dependence  $\Delta C$ .

In conclusion, the most decisive parameter for a good viewing angle dependence of the contrast is  $d\Delta n$ . For a variety of mixtures we were not able to ascertain experimentally a correlation between the elastic or dielectric constants and  $\Delta C$ .



Figure 3. The correlation between  $\Delta \varepsilon / \varepsilon_{\perp}$  and  $\Delta \varepsilon$  for the mixtures of series 1 and 2.

#### 3. Response times

All response time measurements were performed under the same conditions as the determinations of the viewing angle dependence of the contrast. In figure 4, t(on) and t(off), when switched from 0 per cent (just below the threshold) to 90 per cent contrast and from 90 to 0 per cent contrast, respectively, are plotted versus  $d\Delta n$  (series 1). As expected, the response times strongly improve with decreasing  $d\Delta n$ . The same measurements are plotted against the cell gap in figure 5. These diagrams clearly show that the differences between the different mixtures in figure 4 are due mainly to the fact, that a certain  $d\Delta n$  corresponds to a completely different cell gap for the single mixtures. In figure 5 mixtures measured in cells with identical cell gaps show practically the same response times.

In addition we have measured t(on) and t(off) for about 20 mixtures in twisted nematic displays with  $d\Delta n$  of  $0.5 \,\mu m$  (series 2). All mixtures had similar viscosities of around  $20 \,\mathrm{mm^2 \, s^{-1}}$ . However, the  $\Delta n$  values were different, and therefore the cell gaps necessary for  $d\Delta n$  of  $0.5 \,\mu m$  were also different. Again we found that the response times are dominated mainly by the cell gap. This means that again all of the mixtures



Figure 4. The response times t(on) and t(off) as defined in the text versus  $d\Delta n$ . The different symbols represent six different liquid crystal mixtures (series 1, see text).



Figure 5. The response times t(on) and t(off) versus the gap d. The different symbols represent six different liquid crystal mixtures (series 1, see text).

show the same response time for a constant cell gap. The other material parameters, including the elastic constants, exhibit a smaller influence in practice.

From these measurements we find that displays (filled with a liquid crystal optimized for TFT displays) operating in the first minimum with a cell gap of  $6 \mu m$ (corresponding to a  $\Delta n$  of 0.085) typically have t(on) times of about 40–50 ms, when switched from 0 per cent contrast to 90 per cent contrast (at 20°C). This seems to be sufficient for video frame rates. However, the t(on) time increases drastically if the display is switched over a smaller range of the grey scale (whereas t(off) does not change as much). For example, if the  $6 \mu m$  cell is switched from 0 to 30 per cent, t(on)is about 150 ms and when switched from 0 to 50 per cent, t(on) is still approximately 100 ms. On the other hand, a display with a cell gap of  $5 \mu m$  (corresponding to a  $\Delta n$ of 0.1), already requires only about 90 ms for a 30 per cent change in grey scale, about 60 ms for a 50 per cent change, and only about 25 ms for a 90 per cent change in the grey scale (starting from 0 per cent transmission).

The best viscosity values (flow viscosities) at room temperature of practical liquid-crystalline mixtures suitable for TFT displays are between 15 and 20 mm<sup>2</sup> s<sup>-1</sup>. Drastic improvements of the viscosity seem to be practically impossible, if the other properties of the liquid crystal are to be kept unchanged. Therefore, liquid crystals for TFT displays should have  $\Delta n$  values of at least 0.1, suitable for cell gaps of 5  $\mu$ m.

#### 4. Specific resistance

Perhaps the most important parameter of liquid crystal materials for TFT displays is the specific resistance [11–13]. If the resistivity (and hence the *RC* time constant) decreases, the contrast becomes worse and, as the resistivity can decrease with time, this can result in a limited lifetime of the display. Like other organic dielectrics, well purified liquid crystals ought to have very high electrical resistivities. Values of more than  $10^{18} \Omega$ cm are possible. However, in practice, despite careful purification, a residual concentration of ionic impurities remains, resulting in specific resistances of about  $10^{12}-10^{14} \Omega$ cm [14]. In order to have a sufficiently high resistivity over the lifetime of the display, the initial resistivity has to be as high as possible. In addition,



Figure 6. The typical temperature dependence of the resistivity of liquid crystals. The different symbols represent different liquid crystal mixtures.

the resistivity also has to be high at the upper limit of the operating temperature range, e.g. 50 or 60°C (compare figure 6; for the measuring conditions see figure 7). Taking this into account, an initial specific resistance of at least  $10^{12} \Omega$  cm (at 20°C) seems to be necessary for TFT displays. The resistivity of the liquid crystal mixture, of course, depends on the purification of the single compounds after synthesis and (even more) on the optimized purification of the liquid crystal after mixing.



Figure 7. The principle of the resistivity measurement.

The resistivity of the liquid crystal insides the TFT display depends strongly on the purity of the inner surfaces of the display. Here the purity and the polymerization process of the organic polymer, used as the orientation layer, has a distinct influence. However, the most decisive influence comes from the liquid crystal itself. The most important liquid crystal parameter seems to be the polarity or the average dielectric constant  $\bar{\epsilon}$ :

$$\bar{\varepsilon} = (2\varepsilon_{\perp} + \varepsilon_{\parallel})/3$$

This is demonstrated in figure 8, where we have plotted the specific resistance of about 30 liquid crystals, after purification, versus their  $\overline{\epsilon}$ .

As  $\bar{\epsilon}$  is correlated with  $\Delta \epsilon$  for nearly all liquid crystal mixtures of practical interest,  $\bar{\epsilon}$  is also correlated with the threshold. For the same liquid crystal mixture the threshold in a display operating in the first minimum is much lower than for a display operating in the second minimum, or for a certain threshold, a first minimum display requires a liquid crystal mixture with a smaller  $\Delta \epsilon$  and therefore with a smaller  $\bar{\epsilon}$ , resulting in a higher resistivity. Comparing polar and non-polar liquid crystals (large



Figure 8. Resistivity of liquid crystals versus their average dielectric constant  $\bar{\epsilon}$ .

and small  $\bar{e}$ ), we find that: (a) polar mixtures, in general, contain more ionic impurities and are more difficult to purify; (b) polar mixtures can more easily dissolve impurities from the inner display surfaces than non-polar mixtures; and (c) ionic impurities can dissociate much better in polar mixtures (with larger  $\bar{e}$ ) than in non-polar mixtures.

#### 5. Conclusion

We have determined the important parameters of liquid crystals necessary for TFT displays.

- (a) Response times depend in practice mainly on the cell gap, therefore: the display should be operating in the first minimum and the  $\Delta n$  of the mixture should be at least 0.1, corresponding to a cell gap of not more than  $5 \,\mu$ m.
- (b) The viewing angle dependence is influenced by  $d\Delta n$ , and the polarizers orientation, therefore: the display should be operating in the first minimum; the display should have positive constant; and the polarizer should be perpendicular to the liquid crystal director.
- (c) The specific resistance depends on  $\bar{\epsilon}$ , therefore  $\bar{\epsilon}$  must be kept as small as possible (hence the display should be a first minimum display).

Based on these results, we have developed new liquid crystals for active matrix addressing. A few examples are presented in table 2.

Mixture	ZLI-3270	ZLI-3651	ZLI-3771	ZLI-3788	88-680	
Clearing point, $T_{\rm NI}/{}^{\circ}{\rm C}$	91	81	91	90	50	
Viscosity, $v/mm^2 s^{-1}$	20	16	18	18	13	
Optical anisotropy, $\Delta n$	+0.0852	+0.1007	+0.1045	+0.1003	+0.0713	
Dielectric anisotropy, $\Delta \varepsilon$	+5.1	+3.7	+3.7	+4.7	+2.2	
Average epsilon, $\bar{\epsilon}$	5.8	4.8	4.8	5.0	3.6	
Dielectric ratio, $\Delta \varepsilon / \varepsilon_{\perp}$	1.24	1.02	1.02	1.28	0.76	
Threshold, $V_{10}/V$	2.08	2.31	2.44	2.37	2.11	
Elastic constants						
$K_{11}/10^{-12}\mathrm{N}$	13.1	12.8	13.7	13.3		
$K_{22}^{11}/10^{-12}$ N	6.6	7.4	7.0	6.7		
$K_{22}/10^{-12}$ N	18.0	15.5	16.8	18.8		
Specific resistance, $\rho/\Omega$ cm	$2 \times 10^{12}$	$8 \times 10^{12}$	$1.5 \times 10^{13}$	$7 \times 10^{12}$	$1\cdot1 \times 10^{14}$	

Table 2. Newly developed liquid crystal mixtures for active matrix displays.

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