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# Molecular Crystals and Liquid Crystals

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# Multiplexible Liquid Crystalline Broad Range Systems<sup>†</sup>

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A sharp steepness, a low temperature dependence of the threshold, and a fast response time at low temperatures are preconditions for the multiplexibility of liquid crystals suitable for outdoor application. The steepness is efficiently improved if  $K_3/K_1 \le 1$ . A temperature independent steepness and threshold can be realized if  $K_3/K_1$  and  $\Sigma K_1/\Delta \epsilon$  become constant. Response time is proportional to  $\gamma_1/K_1$ . Since viscous properties change by 1 to 2 of the tenth power more than elastic constants with temperature, a fast response behavior at low temperature is possible only by reducing the temperature dependence of the viscosity. Taking into account these preconditions, it is possible to develop nematic systems multiplexible up to 1:8 from  $-20^{\circ}$ C to  $+60^{\circ}$ C without temperature compensation of the driving voltage or with temperature compensation up to 1:16, having a total response time less than 1 s even below  $-20^{\circ}$ C.

#### INTRODUCTION

A liquid crystal suitable for outdoor use, e.g. in an instrument panel, with a maximum driving voltage of 9 volts should still be able to switch within 1 s at a temperature of  $-40^{\circ}$ C and be operational at temperatures of up to at least  $+80^{\circ}$ C. Multiplexibility requires furthermore a steep electro-optical characteristic and a low temperature dependence of the threshold.

Switching time, steepness and threshold are functions of the dielectric, elastic and viscous constants. Consequently a precondition for the development of broad range mixtures is that the temperature dependence of these

<sup>&</sup>lt;sup>†</sup>Presented at the Ninth International Liquid Crystal Conference, Bangalore, December 6-10, 1982.

values and the relationship between the molecular structure and the anisotropic properties of nematic substances are known.

#### **EXPERIMENTAL**

The rise and decay time of a TN (twisted nematic) cell are dependent upon the twist viscosity  $\gamma_1$  and the elastic constants  $K_i$  of the liquid crystal:

$$t_{\text{on}} = \frac{\gamma_1 \cdot d^2}{\pi^2 \cdot K_1(V^2/V_{10}^2 - 1)}$$

$$t_{\text{off}} = \frac{\gamma_1 \cdot d^2}{\pi^2 [K_1 + 1/4(K_3 - 2K_2)]}$$
(1)

where d = display spacing,  $\gamma_1 =$  twist viscosity,  $K_1 =$  splay constant,  $K_2 =$  twist constant,  $K_3 =$  bend constant, V = operating voltage and  $V_{10} =$  threshold voltage.

Values for the temperature dependency of  $\gamma_1$  and  $K_i$  are relatively seldom quoted in the literature. As a result the switching behavior of a liquid crystal is usually characterized by applying the easily attainable flow viscosity.

In order to clarify the question as to what influence the viscous and elastic properties have on the switching times of a liquid crystal at low temperatures and whether it is permissible to substitute the flow viscosity for the twist viscosity, the temperature dependence of the elastic and viscous properties of different liquid crystals with differing polarity in the temperature range from  $-40^{\circ}$ C to  $80^{\circ}$ C was investigated. Some of the physical properties of these nematic mixtures are compiled in Table I.

### **DISCUSSION**

# Viscous and elastic properties

The temperature dependence of the flow and twist viscosity is almost parallel. However as the clearing point is approached the anisotropic twist viscosity becomes zero, while the isotropic flow viscosity retains a final value (Figure 1). With decreasing temperature, the viscosity of liquid crystals with a larger dielectric anisotropy increases greater than for mixtures with smaller  $\Delta \varepsilon$ -values. Over a wide temperature range, the twist viscosity of a liquid crystal is between five to eight times larger than the flow viscosity. Moreover, the stronger polar mixtures incline toward the higher values and the weaker toward the lower values.

TABLE I

Physical properties of broad range liquid crystals with smaller (ZLI-1565, ZLI-1694, ZLI-1957/5) and higher (ZLI-1691, ZLI-1840, ZLI-1982) polarity

	ZLI-1565	ZLI-1694	ZLI-1957/5	ZLI-1691	ZLI-1840	ZLI-1982
sm-nem. [°C]	<-40	<-40	<-40	<-20	<-20	<-20
F <sub>p</sub> [°C]	-20	-20	-16	-5	-15	-15
Clp [°C]	85	86	85	90	90	91
$v*[mm^2s^{-1}]$	19	20	18	33	31	26
$\gamma_1$ *[mm <sup>2</sup> s <sup>-1</sup> ]	130	129	103	258	237	199
$\Delta \varepsilon^*$	6.0	6.5	4.5	14.7	12.2	10.2
$\epsilon_{11}*$	9.9	10.4	8.0	19.4	16.8	14.4
ε <sub>⊥</sub> *	3.9	3.9	3.5	4.7	4.6	4.2
$\Delta n^*$	0.13	0.13	0.13	0.18	0.15	0.14
<i>n</i> <sub> </sub> *	1.630	1.633	1.628	1.679	1.640	1.640
n_*	1.499	1.499	1.500	1.500	1.492	1.497
$K_3/K_1*$	1.3	1.30	0.99	2.10		

<sup>\*</sup>All values for +20°C.

In comparison with the viscous properties, the elastic constants have a relatively small temperature dependence, independent of the polarity of the liquid crystal under investigation.

Accordingly, the viscosities of a purely nematic mixture within a temperature range of 100°C change by a factor of  $10^2-10^3$ ; the elastic constants however change only by a factor of 5 (Figure 2). Switching times at low temperature are consequently determined by the viscous and not by the elastic properties. Because the temperature dependence of the twist and flow viscosity are found to be parallel to each other, it is legitimate to describe the switching times by applying the flow viscosity of a liquid crystal. Under given conditions, the relationship between flow viscosity and response time is found to be largely linear (Figure 3).

Even at temperatures  $10-20^{\circ}\text{C}$  above the nematic-smectic transition, an over proportionately strong increase in the viscosity—due to pretransformational phenomena—deviating from the normal heating pattern is to be observed, and which is also reflected in the switching times. As a result, it is necessary to have a stable nematic phase at temperatures lower than  $-40^{\circ}\text{C}$ . In order to realize fast response times at low temperatures, the determining factor is not that the viscosity at room temperature is extremely small, but that the viscosity has the smallest possible temperature dependence. The precondition for this is a stable nematic phase at temperatures less than  $-40^{\circ}\text{C}$ .

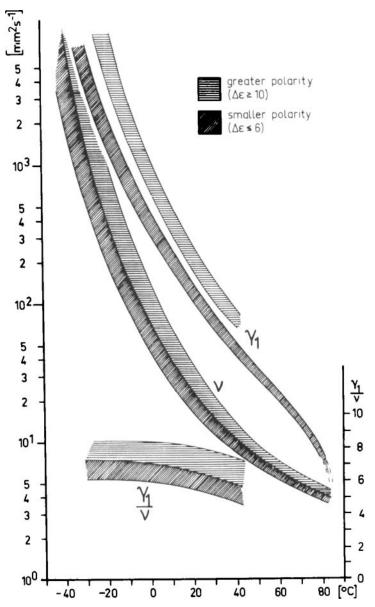


FIGURE 1 Temperature dependence of flow  $(\nu)$  and twist  $(\gamma_1)$  viscosity of liquid crystals with different polarity.

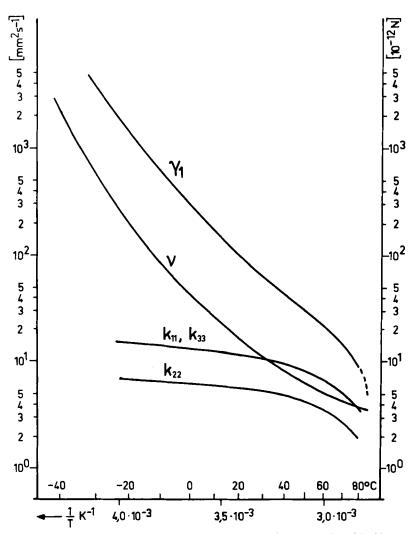


FIGURE 2 Typical temperature dependence of viscous and elastic properties of liquid crystals. Flow  $(\nu)$  and twist  $(\gamma_1)$  viscosity, splay  $(K_1)$ , twist  $(K_2)$  and bend  $(K_3)$  elastic constants of ZLI-1957/5.

# **Dynamic properties**

Liquid crystals become multiplexible if they have a low temperature dependence of the threshold and a sharp steepness of the electro-optical characteristic. According to Alt and Pleshko, the maximum theoretically possible multiplex ratio is given by

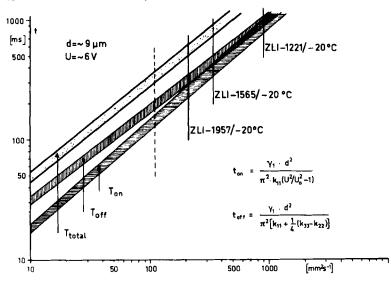


FIGURE 3 Response times as function of the flow viscosity ( $\nu$ ) for various liquid crystals at  $-20^{\circ}$ C. Display spacing of the TN cell: 9  $\mu$ m, driving voltage: 6 volts.

$$N_{\text{max}} = \left(\frac{S^2 + 1}{S^2 - 1}\right)^2 \tag{2}$$

where  $S = V_{90}/V_{10}$  or  $V_{50}/V_{10}$  depending on which contrast ratio is applied. For the threshold  $V_{10}$ , the following known equation is valid:

$$V_{10} = \pi \left[ \frac{K_1 + 1/4(K_3 - 2K_2)}{\varepsilon_0 \cdot \Delta \varepsilon} \right]^{1/2}$$
 (3)

With the aid of the Berreman-Program,<sup>2</sup> Schadt and Gerber<sup>3</sup> have derived the following semiempirical relationship for  $S^* = V_{50}/V_{10}$ 

$$S^* = 1.1330 + 0.0266(K_3/K_1 - 1) + 0.443 \left( \ln \frac{d \cdot \Delta n}{2 \cdot \lambda} \right)^2$$
 (4)

From Eq. (3), it can be seen that a threshold, which is independent of temperature, is given by  $\sum K_1/\Delta \varepsilon = \text{constant}$ . The approximation in Eq. (4) means that a good steepness, that is a small S\*-value, is possible, if  $K_3/K_1 \leq 1$  and if the display spacing and/or the birefringence are so chosen that  $d \cdot \Delta n/2\lambda = 1$ . Under these conditions the second term becomes zero and the first term zero or even negative.

If the transmission characteristic of a twisted cell between parallel polarizers is observed, as was investigated by Gooch and Tarry<sup>4,5</sup> (Figure 4), it is found that an optical retardation  $d \cdot \Delta n/2\lambda = 1$  at a typical wavelength of

# Gooch-Tarry-relation

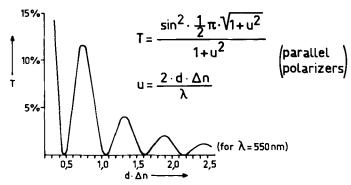


FIGURE 4 Transmission characteristic of a TN cell between parallel polarizers at a typical wavelength  $\lambda$ =550 nm.

 $\lambda = 550$  nm corresponds to exactly the second minimum. Consequently, a precondition for high multiplex ratios is to select cell spacing and birefringence in such a way that the complete electro-optical system operates in the second transmission minimum. In the case that both terms of the mathematical progression in Eq. (4) are zero, a maximum multiplex ratio of 1:64 is calculated from Eq. (2).

#### **RESULTS**

Selecting appropriate components, it is possible to develop broad range mixtures which have not only  $K_3/K_1 \le 1$  but also a  $K_3/K_1$  which is almost independent of temperature (Figure 5). Taking into account these determining conditions, it has been possible to develop liquid crystals which can be multiplexed 1:2 without temperature compensation of the driving voltage within the temperature range  $-20^{\circ}$ C to  $+80^{\circ}$ C, or up to 1:8, again without temperature compensation, within the range  $-20^{\circ}$ C to  $+60^{\circ}$ C, or with temperature compensation, up to even 1:16, in the temperature range  $-20^{\circ}$ C to  $+60^{\circ}$ C. The electro-optical properties of these systems are summarized in Table II.

Provided that a ratio of  $K_3/K_1$  can be achieved which is significantly smaller than 1 and also renders

$$\frac{dK_3/K_1}{dT}=0$$

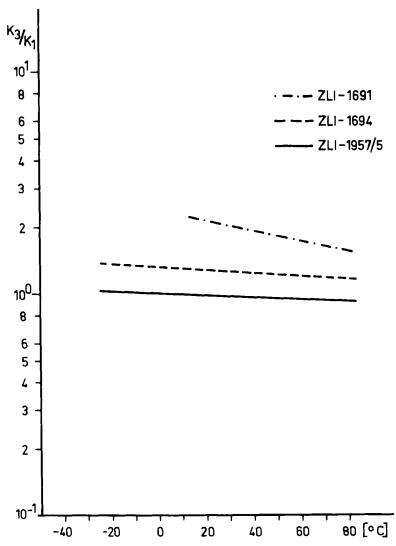


FIGURE 5 Temperature dependence of  $K_3/K_1$  of liquid crystals with different multiplex behavior. Not multiplexible (ZLI-1691); small ratios possible (ZLI-1694); ratios 1:8 possible (ZLI-1957/5).

in this case — on which we are working at present — a furthermore significant improvement in broad range multiplex behavior of liquid crystals is to be expected.

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TABLE II

	Electr	o-optical proper	Electro-optical properties of broad range liquid crystals suitable for multiplex applications	ge liquid crystals	suitable for mu	ltiplex application	SU	
	ZLI-1565	ZLI-1694	ZLI-1957/5	ZLI-2009	ZLI-2116	ZLI-2142	ZLJ-2248	ZLI-2293
sm-nem.[°C] Clp[°C] ν-4αc[mm²s-¹] V <sub>10</sub> [V] Λ(V <sub>10</sub> [V] Λ(V <sub>20</sub> /V <sub>10</sub> ) Λα <sup>2</sup> Γαν Λα <sup>2</sup> Γαν Λα <sup>2</sup> Γαν Λα <sup>2</sup> Γαν	<-40 85 12,300 2.3 8.0 1.16 1.78	<40 86 10,700 2.14 6.3 1.16 1.82 46	<-40 85 2,500 2.6 8.3 1.12 1.60 89 1.16 -20/+60	<40 87 87 5,250 2.1 8.6 1.14 1.78	<pre>&lt;-40 93 4,400 2.5 6.5 1.11 1.69 64 1:2 -20/+80</pre>	<40 85 6,000 2.07 8.0 1.14 1.78	<40 85 7,500 2.17 6.62 1.14 1.76 56	<40 84 6,400 1.94 7.1 7.1 1.14 1.80 56
					i			

 $^*M_{20} = V_{0',20^{\circ}C}/V_{45^{\circ}20^{\circ}}$ 

## **Acknowledgments**

We would like to thank Dr. Hp. Schad (Forschungszentrum of BBC, Baden) for his support in determining  $\gamma_1$  and  $K_i$  values at low temperatures, Prof. Dr. F. Schneider (University of Siegen) for the measurement of the temperature dependence of  $\gamma_1$  and Dr. G. Baur (IAF Freiburg) for his assistance in the evaluation of  $K_i$  values.

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