9.2: Color-Free Plastic LCD with High-Reliability

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A new design has been developed for a plastic substrate twisted nematic LCD. The design results in color free optical performance and high reliability as measured by accelerated life tests at high temperature and humidity. The fundamental design limitation was the crystalline nature of most chemically stable plastics. The crystallinity produces a biaxial birefringence which can produce dramatic interference colors in the twisted nematic geometry. Optically isotropic plastics are generally amorphous and therefore, chemically incompatible with the liquid crystal which is a good organic solvent. We have solved this dichotomy by using a special form of polyester whose optic axes are controlled to lie at an angle from the normal which is considerably larger than the largest angle which can be reached by light entering the high index plastic. We have developed a fabrication process which is consistent with the 150°C glass transition temperature for the polyester as opposed to the 500°C transition temperature of typical LCD glass substrates. We also designed the display materials to be water tolerant, since all plastics trasmit water. We have demonstrated, on a laboratory scale, that this plastic design will pass the Texas Instruments' reliability criteria for high temperature and humidity tests.

Introduction

The twisted nematic (TN) LCD is currently the dominant nonemissive display because of its high contrast and a multiplex capability which, while limited relative to emissive displays, is still better than any other low power display. The need for larger area and higher information content nonemissive displays has been clear for some time, but the cost of new displays has to be reasonable relative to the personal, portable market pulling the technology and relative to the superb cost/performance characteristics of the CRT. We judged that achieving the cost goals for the new markets would be difficult using the standard glass substrate technology which has been so successful in watches and calculators. The film processing capability of the graphic arts industry appeared to offer the potential for the technological jump which would meet these cost goals. Reel-to-reel processing of film substrate LCDs was the goal as discussed by Culley and Surtani ¹¹ in 1980.

Penz, et al.⁽²⁾, of Texas Instruments and Takahashi, et al.⁽³⁾, of Ricoh Watch discussed prototype plastic designs in 1981. Reference 2 discussed a basic materials problem in the fabrication of plastic liquid crystal displays (PLCD), The materials problem is that plastics tend to be either birefringent and chemically robust or isotropic and chemically sensitive. The TN LCD uses polarized light along with a class of liquids which are good organic solvents. In short, substrate materials which are stable with liquid crystals tend to produce strong interference colors while color free PLCD systems tend to be unequal to the environmental tests of many LCD users. Reference 2 discussed the PLCD design shown in Figure 1. Here an isotropic substrate such as cellulose acetate butyrate is 'protected" from the liquid crystal by an abrasion resistant layer of a highly cross-linked silicone material. This design results in devices which are optically equal to or better than glass substrate LCDs. As discussed in Reference 2, however, the water sensitivity of cellulose acetate butyrate leads to significant temperature/ humidity environmental test problems. Abrasion resistant coating designs also lead to a second built-in reliability problem: any pinhole or crack in the resistant layer leads to a catastrophic failure. The Figure 1 design

builds in unreliability and was discarded at TI in favor of a design to be discussed below.

Substrate - Cellulose Acetat	te Butyrate (Isotropic)			
Chemical Resistant I	Layer - Silicone			
Transparent Conductor - ITO				
Alignment	Layer			
Adhesive - Epoxy Li	iquid Crystal - E7			
Alignment Layer				
Transparent Conductor - ITO				
Chemical Resistant I	Layer - Silicone			
Substrate - Cellulose Acetate Butyrate (Isotropic)				
Comments: Chemical Resistant Laver needed to protect				

Comments: Chemical Resistant Layer needed to protect CAB from liquid crystal. CAB fails in humidity test.

Figure 1 Isotropic substrate design for a PLCD along with optical and environmental performance of CAB based design

The birefringent substrate design was discussed in 1981 in both Reference 2 and Reference 3 and is shown in Figure 2. In this design one uses a substrate material such as stretched polyester. The problem with a standard polyester substrate is its biaxial birefringent characteristic as discussed in detail by Umeda, et al.⁽⁴⁾, in 1982. The birefringent character results from the polymer crystallinity induced by the stretching process, which also produces the chemical robustness by inducing chemical bonding between the polymer claims. Figure 2 is the basic design discussed in Reference 3, but the birefringence color problem was not discussed in Reference 3.

Substrate - Biaxial Draw Polyester (Mylar) **Transparent Conductor - ITO** Alignment Layer - Polyvinyl Alcohol Adhesive - Epoxy Liquid Crystal - E7 Alignment Layer - Polyvinyl Alcohol **Transparent Conductor - ITO** Substrate - Biaxial Draw Polyester (Mylar) Comments: Polyester stable with liquid crystal. Biaxial substrate led to severe birefringent colors. Figure 2 Biaxial draw polyester design for a PLCD Substrate The technological key to achieving this report of a color free, high reliability PLCD is the use of a polyester

substrate which is fabricated so as to have optic axes which are at a sufficiently large angle relative to the substrate normal so as not to interfere with light incident on the substrate from the outside air. A very useful technique for experimentally investigating the

birefringent properties of a LCD in general and a PLCD substrate in particular is convergent light microscopy. The conoscopic figure of a standard piece of Mylar (trade name for duPont polyester film stretched in two orthogonal directions) is shown in Figure 3. The picture was taken usng a 50x/NA = .85 objective lens, a NA = .9 condensing lens and sodium light. The optic axes, or metalopes, are the bull's-eye figures outlined by the sodium interference fringes. The edge of the conoscopic disk corresponds to an angle of $\sin^{-1.85} = 58^\circ$, whereas the optic axes lie at an angle of approximately $\beta = 28^{\circ}$ as observed in air. The conoscopic figure was taken between crossed polarizers oriented for extinction relative to the plastic substrate. Regions which are dark in the figure do not depolarize the light whereas bright regions do. In the mylar substrate itself, the optic axes lie at an angle a =18°. Light incident on the substrate from the air can propagate at angles up to $\Delta = \sin^{-1} 1/1.5 = 42^{\circ}$. It is clear, therefore, that this substrate will produce strong interference colors, even at an extinction orientation of the polarizers.





Figure 3 Conoscopic figure (NA = .85) and cross sectional view of a biaxial draw sample of polyester. The optic axes are at $\alpha = 18^{\circ}$ in the plastic and appear at $\beta = 28^{\circ}$ in air. The angles greater than $\Delta = 42^{\circ}$ are blocked out because light from the air cannot achieve these angles by normal refraction for an index of 1.5. In the bright regions of the conoscopic figure, light is depolarized and one will see color in a PLCD due to the depolarization.

It is possible to prepare polyester substrates which are biaxial but whose optic axes lie at a much larger angle from the normal than with Mylar. The technique employed is to stretch the polyester film in only one direction. Figure 4 shows the conoscopic figures which result from uniaxial stretch (UAS) vs. biaxial stretch (BAS) polyester films using an oil emergence conoscopic technique. The numerical aperture of the emersion lens

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was 1.32. The oil emergence technique was used because a standard air medium lens technique will not detect the optic axes of the UAS film — which also means no interference colors in a PLCD. In Figure 4 the edges of the disks correspond to 62° in the film itself. The UAS optic axes are at $x = 51^\circ$ and the BAS optic axes are at $x = 18^\circ$ from the normal. The superior performance of polarizers made using UAS polyester as protective layers is discussed in Reference 6.



BIAXIAL PET



UNIAXIAL PET

Figure 4

Conoscopic figures taken with an oil emersion microscope (NA = 1.32) using BAS and UAS pieces of polyester. The optic axes of the UAS material lie at 51° which is larger than the 42° maximum for refracted incident light.

The PLCD is fabricated using the UAS film as follows. First, one prepares the electrode patterns on the two substrates so that the optic axes of the substrates are parallel to each other. This is necessary to match the expansion coefficients of the anisotropic substrates and avoid warping of the finished laminated cell. The second step is to prepare alignment layers which are accurately parallel to the optic axes projection on the substrate on one substrate and perpendicular to the optic axes projection on the other substrate in order to achieve the twisted nematic geometry. Registration of the liquid crystal alignment direction and the substrate optic axes projection to better than $\pm 2^{\circ}$ is necessary to avoid depolarization in the substrates and consequent loss of contrast. Naturally, one also takes care to orient the polarizers with equal accuracy relative to the substrate optic axes projection. The new design is shown in Figure 5 and will be the basis for the rest of the discussion concerning reliability performance. Reliability and performance data of Figures 1, 2, and 5 are given in Table 1.

Substrate - Uniaxial Draw Polyester					
Thin Film Insulator					
Transparent Conductor - ITO					
Alignment Layer					
Adhesive - Epoxy	Liquid Crystal - 653				
Alignment Layer					
Transparent Conductor - ITO					
Thin Film Insulator					
Substrate Uniaxial Draw Polyester					

Figure 5 Uniaxial draw polyester design for a PLCD

Table 1

Display Performance Test:	s Figure 1	Figure 2	Figure 5
Threshold Voltage Color Free Viewing Good Contrast All Segments Operate	Pass Pass Pass Pass	Pass Fail Pass Fail	Pass Pass Pass Pass
Display Reliability Tests	Figure 1	Figure 2	Figure 5
Temp. Storage High Humidity Operating Life Temperature Cycle	Fail Fail	Pass Pass	Pass Pass Pass Pass

Thin Film Considerations

The second major problem associated with the PLCD fabrication process is the deposition of crack free thin films, especially the transparent conductor of indium tin oxide (ITO). The ITO coating must be deposited with reasonable sheet resistance, $100-1000\Omega/\Box$, using processing temperatures which are consistent with the polyester crystalline polymer, typically temperatures less than 150°C. Magnetron sputtering techniques can be used to successfully deposit ITO films under such conditions. There is also the question of adhesion between the ceramic ITO layer and the much softer polymer which has been specifically fabricated to be chemically inert. The standard processing prescription to improve adhesion is to preprocess the surface by plasma etching. Presumably this cleans, chemically activates, and hardens the polymer surface. Rolls of polyester film often have a slip agent added to one side of the film to keep the adjacent layers of the film from sticking together. We used the clean, pure side of the film as the LCD processing side to avoid contamination from the slip agent. We have found it possible to produce very tough ITO films on polyester by such methods.

Many glass LCD processes call for the addition of a thin film over the glass to prevent ions from migrating from the glass into the liquid crystal. Figure 5 indicates that such a blocking layer is useful for PLCD processing. Typical polyester film processes result in short chain polymers which are very mobile and can constitute up to 2% of the film by weight. We have found that a 1000 Å blocking layer of a ceramic like Al_20_3 has important implications in the high temperature (high humidity) environmental tests. Specifically, we have found that stress for the blocking layer design than for the bare polyester design. Presumably this is due to the short chain polymers being kept out of the liquid crystal with the blocking layer design.

Alignment Layer and Liquid Crystal Materials

The first and foremost consideration for the alignment layer and the liquid crystal material in a PLCD is the fact that the substrates transmit water and so a hermetic seal is impossible. Fortunately, liquid crystal materials which are water tolerant have been available for some time. We used a commercial mixture Roche 653 because we had large quantities of the material on hand and because the component materials, mainly Demos, esters, and pyrimidines, are known to be water tolerant. We used a small percentage of BDH C15 as a chiral additive, typically 0.1% by weight. We found water sensitive materials such as Shiff base structures to be unacceptable due to hydrolysis.

The question of the alignment layer is a tougher problem. The well known polyimide polymerization at 450°C which works so well for a glass process obviously is unsuitable for a PLCD process. We developed a water tolerant alignment layer with much lower processing temperatures than polyimide. This layer will not be discussed further here. See Reference 7 for another discussion of alignment layers for a PLCD.

Perimeter and Fill Hole Seal Considerations

The adhesive technology used to seal the two substrates together is another consideration in a PLCD process that is different from a glass process. Again, high temperatures can not be used, and one would like to have a very rapid seal process to be consistent with the reel-toreel design goals for the PLCD process. We were able to achieve a low temperature seal using a low outgassing epoxy. We did not, however, develop the rapid seal technology. This paper reports on a research effort whose goal was to demonstrate that a batch process, hopefully but not necessarily consistent with a reel-to-reel process, could produce devices with high reliability performance in environmental tests. An interesting approach to the reelto-reel seal process has been disclosed in a patent by Umeda, et al.⁽⁸⁾

The fill hole seal process employed for the PLCD work is unique to a plastic substrate. We found that it was possible to weld the fill holes shut using polyester on a hot carrier -- in the presence of liquid crystal material. The welded seal was clearly an amorphous polyester weld and therefore, not a high quality seal. We followed the polyester weld with an epoxy adhesive which could be quite reactive since the amorphous polyester separated the liquid crystal from the epoxy and its reactive products.

Environmental Tests

The most difficult environmental tests during the PLCD development work were the high temperature, high humidity tests due to the close proximity of the test temperatures to the characteristic temperatures of the plastics, e.g., the glass transition temperature. The major result of this report is the performance of the PLCD process described above under the following three tests: (1) 70°C at ambient humidity for 1000 hours, (2) 50°C at 95% relative humidity for 360 hours, and (3) 50°C at ambient humidity with 3V RMS applied for 1000 hours. The 70°C test was performed with a daily temperature profile of 70°C for 16 hours and room temperature for the remaining 8 hours until the 1000 hours at 70°C had been achieved. This temperature profile was thought to be a closer approximation to an unregulated warehouse environment. The criterion for passing the test was that the current drawn by the display should not have increased by more than 50% as measured 24 hours after the displays had been removed from the environmental chambers.

One strategy which we found very useful to successfully meet these very stringent environmental tests was to adjust the processing conditions to achieve the minimum resistive current, IR, relative to the capacitive current. I. Using the ratio of $I_{c'}I_R = D$, as measured by a phase sensitive technique, we adjusted the process such that the initial ratio was 4 or greater. Since the measured RMS current I is equal to $I_c(1 + (I_R/I_C)^2)^{1/2}$ and since the normal change in current under environmental tests is for IR to increase whicle I_c remains the same, starting with a maximum I_c/I_R is a distinct advantage.

The geometrical format used to conduct the environmental tests was a cell approximately one square inch in total area and a nominal gap thickness of $13 \,\mu m$. The results of the tests are shown in Tables 2, 3 and 4. The current increase in all three tests is less than the 50% specification. These tests are very rigorous and cannot be met by many polarizers used in TN LCDs. We believe that these tests establish the technical feasibility for a reliable. color free PLCD.

Table 2 70°C TEST*/1000 HOURS

Cell	D(t = 0)	D(t = 1000)	I(t=0)	I(t = 100)	0) Δ I/I
450	3.8	2.9	672	730	9%
452	3.1	1.6	651	777	19%
454	5.0	3.8	673	754	12%
456	4.3	2.2	708	823	16%
457	3.2	1.7	503	641	27%
*NOTE	: Sp	ecification is 2	I/I < 5	0% at 1000	hours
CONCL	USION:	Pass Test			

Average Current Increase = 17%

Table 3 50/95 TEST*/432 HOURS

Cell	D(t=0)	D(t = 432)	I(t=0)	I(t = 43)	2) 🛆 I/I	
464	4.3	1.0	575	856	49%	
465	5.0	1.2	621	839	35%	
466	Fail - O	pen				
467	Fail - S	hort				
469	4.0	2.1	925	1051	14%	
471	4.2	2.2	820	854	6%	
472	4.2	2.0	698	888	27%	
*NOTE	: Spe	ecification is	$\Delta I/I < 50$	% at 360	hours	
CONCI	LUSION:	Pass Test				
		A			000	

Average Current Increase = 26%

Table 4 50°C @ 3VTEST*/1000 HOURS

Cell	D(t=0)	D(t = 1000)	I(t = 0)	I(t = 1000)	I/I 2 ((
460	5.7	4.0	578	618	7%
461	4.4	3.6	686	715	4%
462	6.2	4.3	698	759	9%
463	.6.7	3.9	475	526	11%
*NOTE	E: Sp	ecification is .	1/I < 50)% at 1000	hours
CONC	LUSION:	Pass Test	•		

Average Current Increase = 8%

Conclusion

We have demonstrated a laboratory scale design for a PLCD which is capable of excellent performance in environmental tests for reliability. These results were achieved by the use of a novel form of polyester which has both the optical and chemical characteristics to make a reliable PLCD. A PLCD process was developed to be consistent with the low glass transition temperature of the polyester relative to the float glass used in glass substrate LCDs and to be consistent with the water permeability of the polyester substrate. The laboratory process is generally consistent with reel-to-reel film processing with the exception of the perimeter seal technology. We are confident that further work can solve this problem, e.g., Reference 8. We feel that this work has shown one design for a plastic LCD to be technically feasible with consumer type reliability. One could expect to see a production version of a PLCD some time in 1984. It will be interesting to see if the advantages forecast for the plastic LCD will, in fact, enable a plastic substrate design to successfully compete with the well established glass process.

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Bibliography

- B.G. Culley and K. Surtani, U.S. Patent 4,228,574 1) (1980)
- P.A. Penz, K. Surtani, W.Y. Wen, M.R. Johnson, 2) D.W. Kane, L.W. Sanders, B.G. Culley, and J.G. Fish, SID Symp. Dig. Tech. Papers 1981, p. 116. S. Takahashi, O. Shimokawa, H. Inoue, K. Uehara,
- 3) T. Hirose, and A. Kilyama, SID Symp. Dig. Tech.
- T. Umeda, T. Miyashita, and F. Nakano, SID Symp.
 Dig. Tech. Papers 1982, p. 178.
 P.A. Penz, Proc. SID, 19 #2, 43 (1978). 4)
- 5)
- L. Bolt, SPIE 307, 22 (1981). 6)
- 7) H. Inove and A. Kikuyama, Proc. First Euro. Disp. Res. Cont, p. 28 (1981). T. Umeda, T. Igawa, Y. Simazaki, T.Miyashita, and
- 8) F. Nakaus, U.S. Patent 4,362,771 (1982).