Performance Degradation of OMEGA Liquid Crystal Polarizers

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1. Abstract

Liquid crystal polarizers (LCPs) are used in OMEGA to control circularly polarized light due to their unique optical property of selective reflection, whereby light of a specific wavelength and handedness is reflected. This mechanism also acts as a safety feature to prevent potentially damaging back-reflections in OMEGA. It was recently discovered that the selective reflection wavelengths of approximately 42% of the LCPs have shifted by more than 15 nm toward both bluer (shorter) and redder (longer) wavelengths, rendering 21% largely or completely ineffective. Research focused on determining if a mechanical effect could cause these wavelength shifts. Experiments were carried out which demonstrated that a compression or expansion of the gap between the substrates could result in either a blue shift or a red shift, respectively, of the selective reflection wavelength. These results support the hypothesis that a change in the gap thickness of an LCP device could result in a distortion of the pitch of the liquid crystal, which in turn shifts the selective reflection wavelength and degrades the LCP performance.

2. Introduction

2.1. Chiral Nematic Liquid Crystals

The molecules in a crystal possess orientational and positional order, while no molecular order is found in a liquid. There is a state of matter which exhibits the characteristics of both liquids and crystals known as a liquid crystal (LC). The most basic LC is a nematic, which exhibits only orientational order. A *chiral* nematic LC (Fig. 1) is created by adding a twisting agent to a nematic mixture. In the chiral nematic phase, the alignment of the molecules in each layer is rotated with respect to the previous layer.

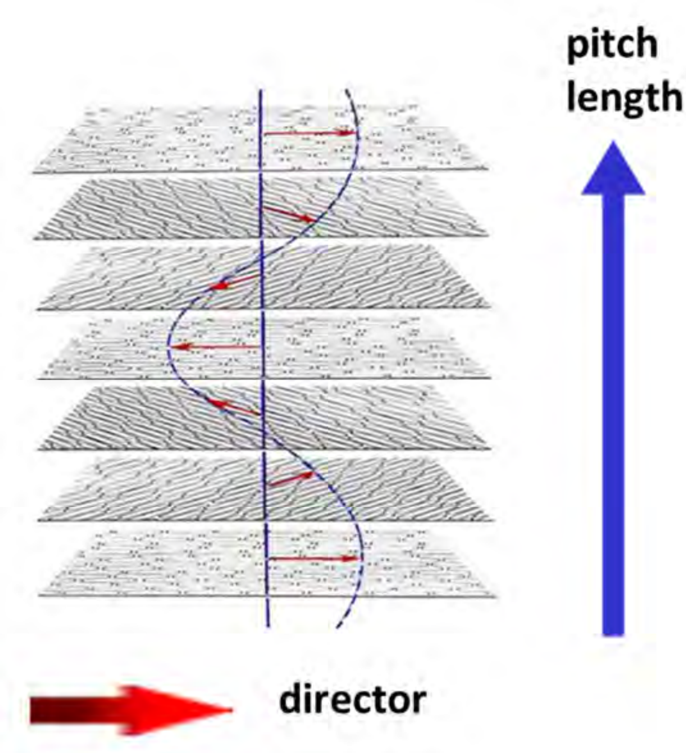


Figure 1: A depiction of a chiral nematic LC. The director denotes the average orientation of the molecules in each layer. The pitch length is the distance required for the director to make a 360° rotation.

This molecular structure leads to the selective reflection of circularly polarized light of a specific wavelength and handedness. Circularly polarized light with the same handedness as the chiral nematic helix will pass through the LC, while light with the opposite handedness will be reflected. A simplified version of Ferguson's equation[1] (Eq. 1) gives the central wavelength (λ_c) for a normal angle of incidence and reflection (See Appendix A):

$$\lambda_c = n \bullet p \qquad \text{(Eq. 1)}$$

where p is the pitch length and n is the mean refractive index of the LC.

2.2. Liquid Crystal Polarizers (LCPs)

Almost 200 LCPs are used in OMEGA to control the polarization of the laser light (Fig. 2). As a laser beam propagates through various optical components, its polarization state can be altered. On OMEGA, most LCPs are right-handed circular polarizers (CPRs), which restore the proper polarization by reflecting the left-hand circularly polarized components of the light. Propagating properly polarized light improves energy balance between beamlines, but more importantly it acts as a safety feature by preventing light from propagating backwards through the system. The CPRs in the OMEGA system are placed at a 10° angle of incidence so that the reflected light is removed from the system. The CPRs are designed to reflect light at 1053 nm at this placement angle; when they are measured at normal incidence the λ_c red-shifts by about 7 nm to 1060 nm.

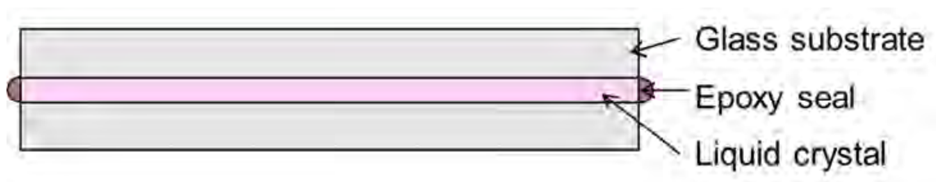


Figure 2. LCPs consist of two glass substrates, a thin layer of chiral nematic LC fluid, and an epoxy seal. The LCPs are filled at an elevated temperature, cooled to room temperature, sheared, and sealed.

The LC mixture used in the CPRs consists of 19.44 %wt CB15 and 80.56 %wt ZLI 1167. The outside surfaces of the substrates, which are made of BK-7 and fused silica, have AR coating. The inside surface of each substrate has three thin film spacers outside the clear aperture resulting in an 18 micron gap.[2]

2.3. Discovery of the degradation and initial data

In October 2011, it was noted that the contrast ratios of several sets of crossed LCPs (a CPR and a left-handed circular polarizer, or CPL) in the OMEGA laser system were less than 100:1, much lower than the specified 1000:1. Three of the four CPRs scanned in the Lambda-900 Spectrophotometer (see Section 3) showed a λ_c which had shifted away from the desired λ_c and towards the longer (red) wavelengths. Due to this discovery, a portable spectrometer was designed and used to perform *in-situ* characterization of 172 CPRs in the OMEGA laser bay. Surprisingly, these measurements found that while many CPRs had a red-shifted λ_c, the selective reflection of some CPRs had shifted towards shorter (blue) wavelengths (Fig. 3). This comprehensive survey found that 42% of the tested CPRs were ineffective polarizers at 1053 nm.[3]

Performance degradation due to the spectral shift of λ_c has not been correlated to the age, location, size, or LC batch of the CPRs. Note that none of the four CPLs that were measured have exhibited a spectral shift.

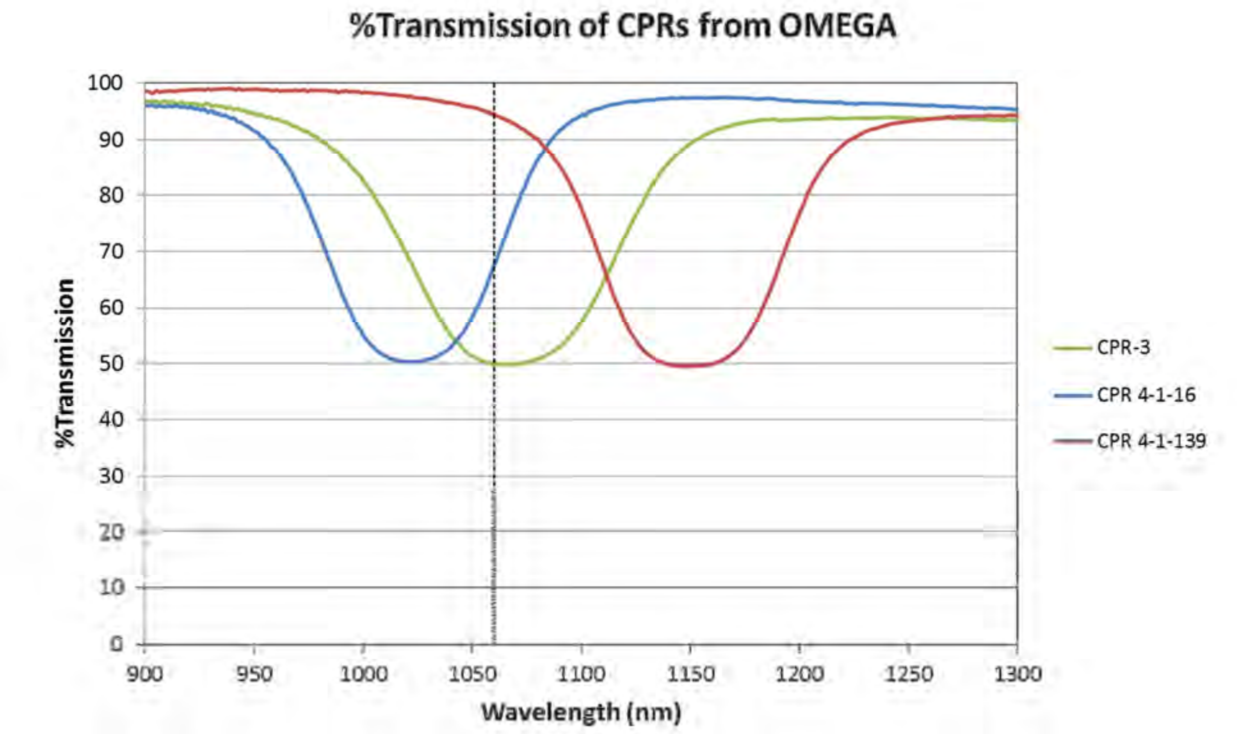


Figure 3. The percent transmission (%T) of three CPRs from the OMEGA system. Because the spectrophotometer emits equal amounts of right and left circularly polarized light, 50% transmission indicates that the CPRs are reflecting the correct amount of light. The dotted line indicates the specified λ_c at normal incidence (1060 nm). CPR-3 is performing well, but CPR 4-1-16 and CPR 4-1-139 have shifted to the blue and red, respectively, and are ineffective polarizers at 1060 nm.

2.4. Possible Cause of Degradation

Many factors can influence and change the selective reflection of LCPs, including temperature, variance in the chemical composition of the LC batches, chemical reactions, and mechanical deformation. The first two factors can be eliminated: the LC mixture used in the CPRs was chosen for its minimal temperature sensitivity and LC batches were found to be consistent with each other. The possibility of either chemical or mechanical change must still be investigated. Assuming that the temperature dependence of the refractive index of the LC fluid is negligible, Eq. 1 suggests that the spectral shift is due to a change in pitch length. Thus, factors that change the pitch length must be examined.

A hypothesis proposing a mechanical origin for changes in the selective reflection was constructed based on an article by Bailey *et al*,[4] in which Bailey described an electrically induced distortion of substrates which led to a change in the pitch length. In the case of OMEGA CPRs, a mechanically induced pitch distortion causing the LCP degradation is a plausible mechanism which could produce either a red or a blue shift. This hypothesis also supports the lack of correlation with device age, device location in the laser bay or storage, and LC batches.

Assuming that there is nowhere for the LC fluid to flow and redistribute itself, a change in gap thickness would require that the spacing between the molecular layers changes as well, thereby altering the effective pitch length. In theory, for a material with n = 1.57 and a λ_c = 1060 nm, a gap thickness change of ~ 0.5 microns, or a pitch length change of about 2.8%, corresponds to a 30 nm shift to the red or blue (see Appendix B).

3. Experimental

The main goal of this project was to investigate the possibility of mechanical degradation of the CPRs by studying the relation between gap thickness and λ_c. Devices were characterized and the central wavelengths of the CPRs were monitored during all experimentation using the Perkin-Elmer Lambda-900 Spectrophotometer, which measures the percent transmission (%T) of samples across a spectrum of wavelengths.

Two sets of CPRs were investigated. The first was a set of "old" CPRs that were stored in a drawer in the Materials Lab for approximately 15 years. The age, LC fluid composition, substrate type, type and thickness of the spacers, epoxy composition, and coating of the CPRs are unknown (see Appendix C). The second was a set of "new" CPRs removed from the OMEGA system because they were no longer effective at 1053 nm and needed to be replaced.

3.1. Characterization and study of "old" and "new" devices

The "old" CPRs were scanned in the Lambda-900 from 900 to 1300 nm to obtain the %T and λ_c of each CPR. Each CPR was scanned at the center of its clear aperture, and each was found to be red-shifted. The "new" CPRs, whose λ_c had already been determined *in situ* in the Laser Bay, were scanned from 800 to 1500 nm at ½ cm intervals across their diameters to assess the uniformity of device performance. In some CPRs the λ_c was fairly uniform across the device, while in others the λ_c varied by as much as 14 nm across the device. This variation could suggest that the gap is not uniform across the diameter (Fig. 4), but results are not conclusive. Note that four of these "new" CPRs are blue-shifted and the other five are red-shifted. So far, no correlation has been found between the direction of spectral shift and the variations across the diameter. This data was used as a reference in later experiments (see Appendix C).

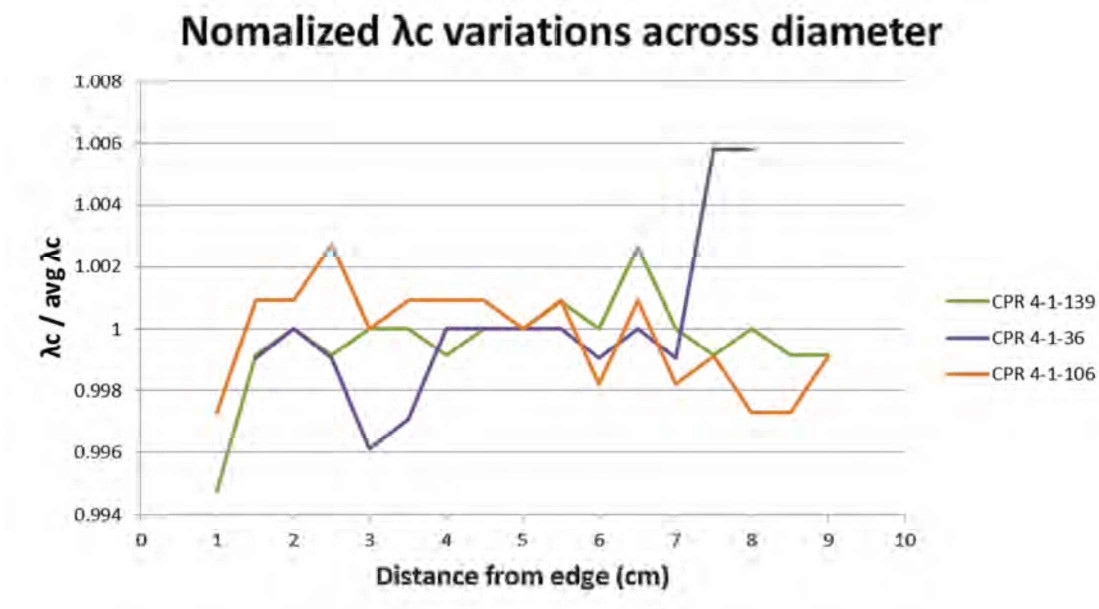


Figure 4. The central wavelengths, λ_c, of three of the scanned CPRs. There are variations in λ_c across the diameters of these CPRs.

3.2 Inducing a change in selective reflection

Several experiments were devised in order to study the effect on λ_c of increasing or decreasing gap thickness. The temperatures of the CPRs and the Lambda-900 testing chamber were not controlled over the duration of these tests. Experiments had been conducted to verify that small temperature fluctuations do not affect the λ_c.

3.2.1 Wedged Cell: Comparing gap thickness and λ_c

A wedged cell (Cell-104) was made by setting a small piece of 12.3 micron Mylar spacer on one end of a microscope slide, placing a line of OMEGA CPR LC fluid parallel to the length of the slide, and dropping another slide on top. Epoxy (5-minute by Devcon) was applied and allowed to cure. The cell was then scanned in the Lambda-900 at 5-mm intervals from the Mylar spacer along the length of the cell. A correlation was observed between the gap thickness of the cell and the λ_c (Fig. 5); as the gap thickness increased, so did the λ_c.

It is important to note that while this cell assembly was performed at room temperature, the OMEGA CPRs are assembled at an elevated temperature with LC fluid in the isotropic phase. Filling the CPRs in the isotropic phase allows the LC to be aligned more easily; however, filling in the isotropic phase is not required to align the LC fluid.

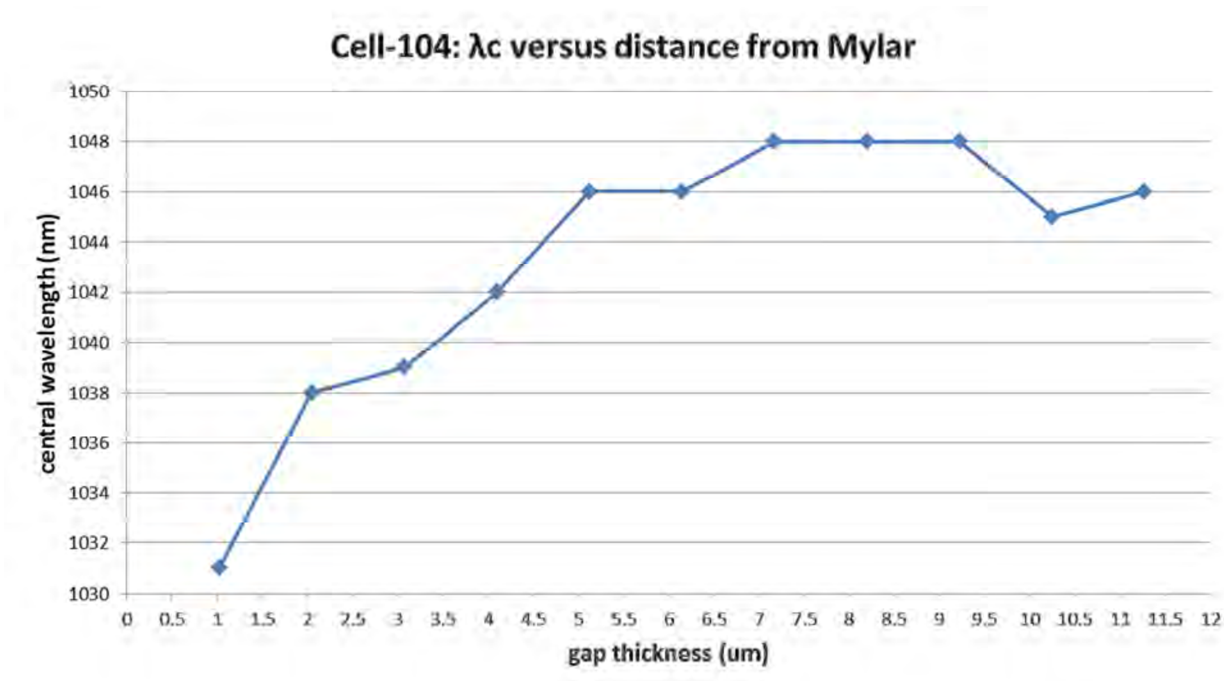


Figure 5. Central wavelength, λ_c, of a wedged cell (Cell-104) as a function of gap thickness. As the distance from the spacer increased, the gap thickness decreased, and the λ_c decreased.

3.2.2 Compressing the device gap

Using a small C-clamp, pressure was applied to CPR-730 and CPR-132 in several separate tests to study the relationship between changing gap thickness and λ_c.

During each test, rubber was placed on the ends of the clamp to protect the substrates. The edge of the clamp was lined up with the fiducial mark on each CPR, placed 1 inch in from the edge of the CPR, and tightened just enough to grasp the device. The device under test (with the clamp) was placed in the Lambda-900 (Fig. 6(b)). The clamp was tightened at one-quarter turn intervals, and the %T was measured at each interval using the Lambda-900. The total clamp forces were quantified using a Shimpo Digital Force Gauge (Fig. 6(a)). During the second and third tests on CPR-132, the CPR was placed in a holder to prevent it from rolling sideways or tilting relative to the incident light, thereby increasing the accuracy of the transmission scans. The CPR-730 tests were performed within one week of each other, as were the CPR-132 tests. For each CPR, the clamp was tightened and loosened three times, and the CPRs were scanned in approximately the same location each time.

Number of quarter turns	Approx. newtons applied
1	9
2	28
3	55
4	87
5	115
6	183
7	213 (estimated)
8	240 (estimated)

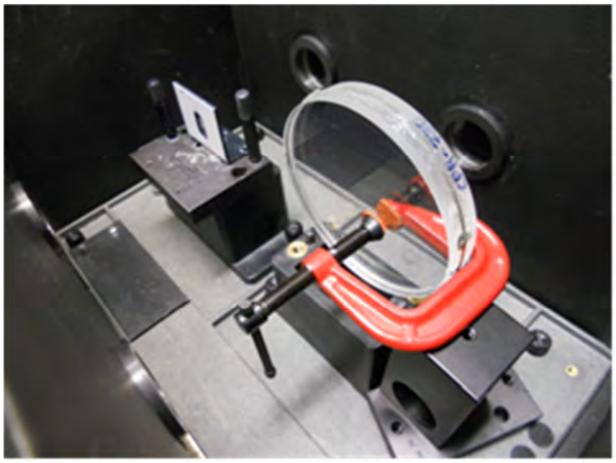


Figure 6. (a) Approximate newtons applied to the CPRs. (b) CPR-730 and clamp in the Lambda-900 testing chamber.

The tests on CPR-730 (Fig. 7) suggest that there is a correlation between λ_c and applied pressure – as the pressure increased, the λ_c decreased. The incident light was approximately normal to the device, and the λ_c began to shift towards the blue when the applied pressure reached a threshold of about 60 newtons. The λ_c of CPR-132 did not show the same dependence on applied pressure (Fig. 8).

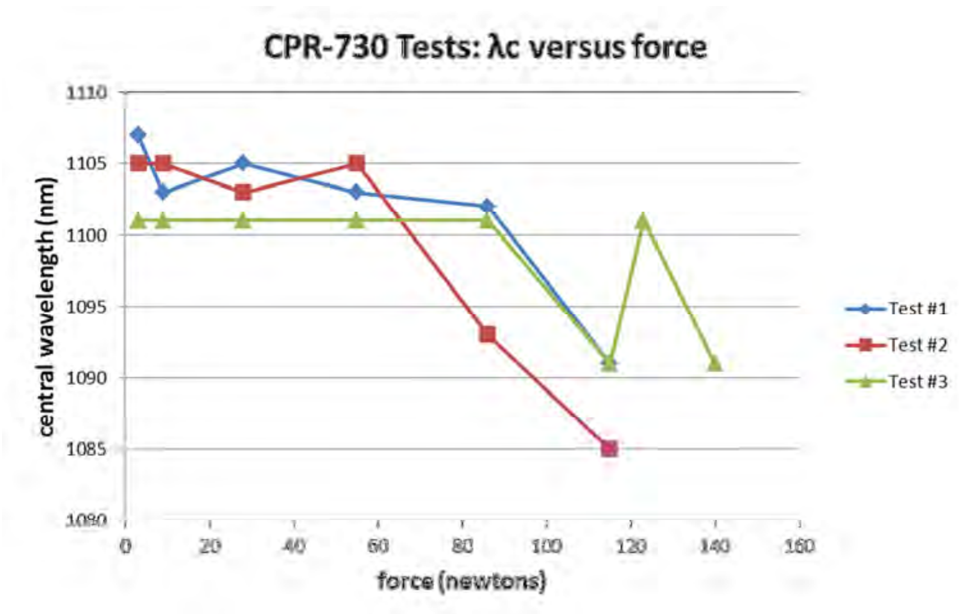


Figure 7. The correlation between λ_c and force applied to CPR-730 for all three pressure tests. It seems that as pressure was increased, the gap thickness and λ_c decreased. It is believed that the outlier at ~125 N was caused by inaccuracy of the transmission scan.

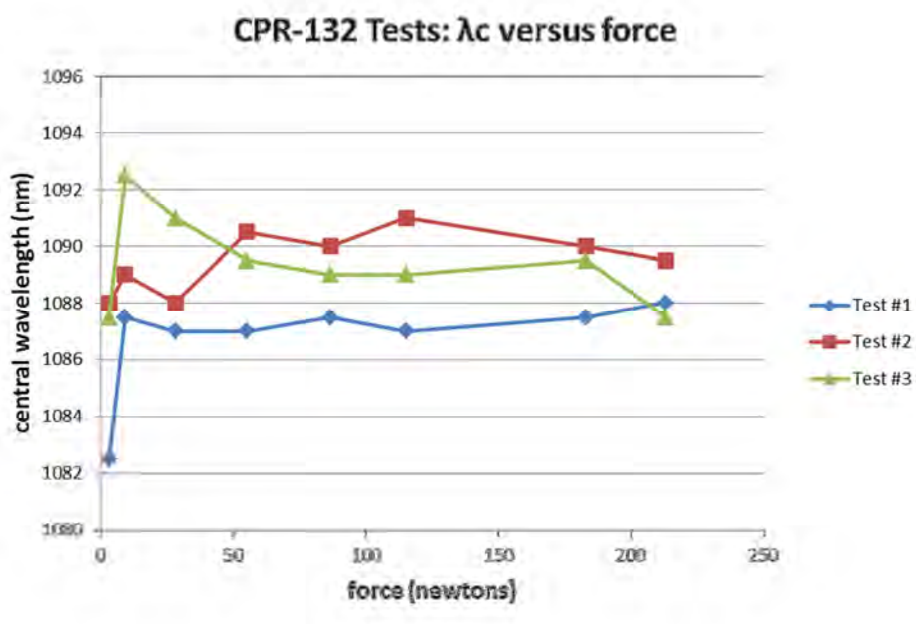


Figure 8. The central wavelengths and force applied to CPR-132 for all three pressure tests. In contrast to Figure 7, no clear correlation is seen between applied pressure and λ_c.

The central wavelengths of CPR-730 and CPR-132 were found according to the equation

$$\lambda_c = \frac{h_1 + h_2}{2} \qquad \text{(Eq. 2)}$$

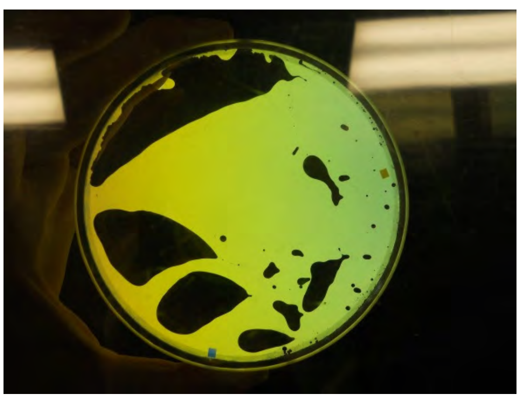
where h_1 is the estimated half maximum on the left side of the %T curve and h_2 is the estimated half maximum on the right side of the %T curve.

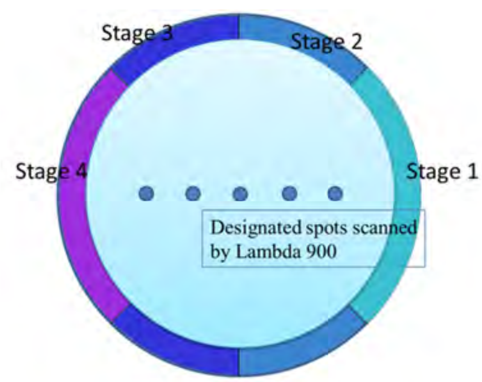
The results of the tests on CPR-730 show a definite correlation between applied pressure and λ_c. However, the results of the tests on CPR-132 do not necessarily show a correlation. Additional testing needs to be carried out to more fully understand these results.

3.2.3. Expanding the Device Gap to Increase λ_c

This experiment was instrumental in the development of a procedure for disassembling CPRs. (See Appendix D for full procedure.)

Before any epoxy was removed, CPR-443 was scanned in the Lambda-900 across its diameter in five spots designated by a mask (Fig. 9(b)). The epoxy was then removed in stages (Fig. 9(b)) using a razor blade and pick. After each stage was removed, CPR-443 was scanned in the same five spots. Air bubbles entered the CPR when the first stage of epoxy was removed, and the bubbles continued to migrate throughout the CPR over the course of the experiment (Fig. 9(a)). Once the CPR had equilibrated for several days, it was again scanned in the Lambda-900 to obtain the equilibrium λ_c.





(a) **(b)**

Figure 9. (a) CPR-443 viewed between crossed polarizers after 50% epoxy removal. Note that the dark areas are air bubbles and the LCs appear yellow/green. (b) Depiction of spots scanned by Lambda-900 and epoxy-removal stages of CPR-443.

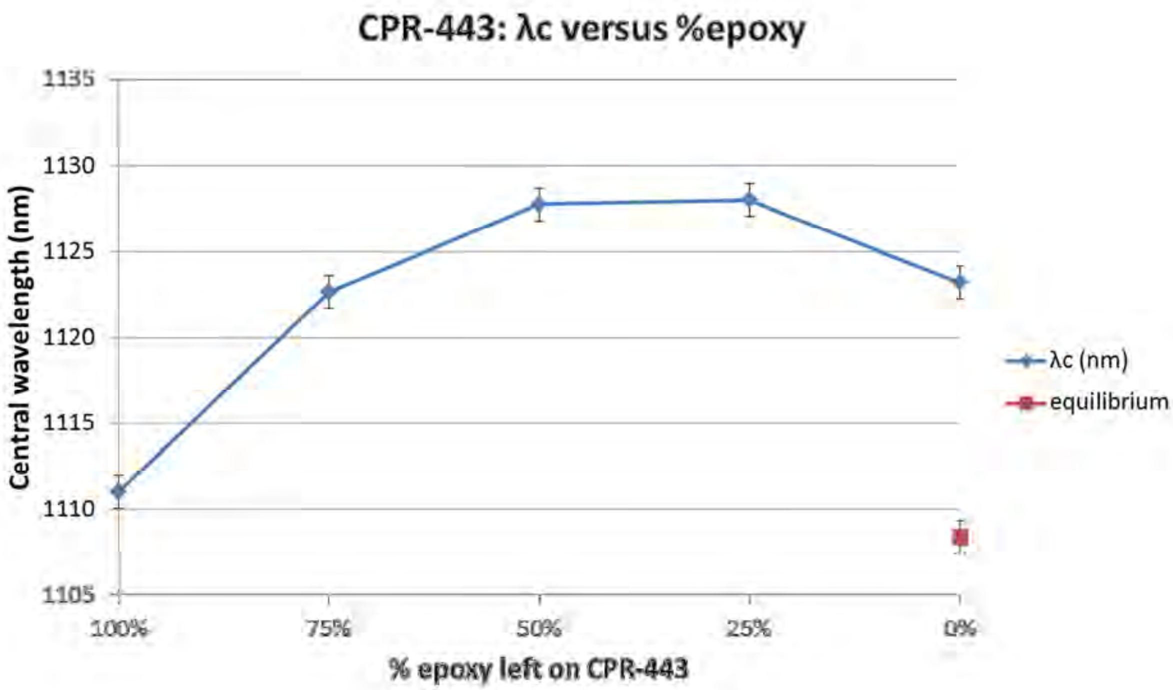


Figure 10. The λ_c increased as epoxy was removed and air bubbles entered the gap between the substrates.

The experiments described in Section 3.2.2 dealt with decreasing the gap thickness, whereas this experiment increased the gap thickness. The results are shown in Fig. 10. As more epoxy was removed and additional air bubbles entered the gap between the substrates, the λ_c increased from its value measured at the onset of disassembly until all the epoxy was removed. These results suggest that the bubbles pushed the substrates apart and increased the gap thickness, which led to an increase in λ_c. Final scans showed that once the air bubbles and LC fluid distribution equilibrated, the λ_c returned to the value recorded at the start of the experiment, which in turn suggests that the pitch length returned to its value from the beginning of the experiment.

Before the experiment began, the λ_c of CPR-443 was measured to be 1111 nm. If it is assumed that the proposed hypothesis is correct, and that the original λ_c of CPR-443 was 1060 nm, it is not clear why the λ_c only returned to 1108 nm when the epoxy was removed. It is possible that the hydrostatic pressure of the LC fluid prevented the gap thickness from changing further, or that a chemical change in the LC fluid has occurred.

4. Conclusions

Various experiments were carried out to investigate the hypothesis that a mechanically induced pitch distortion is causing the LCP performance degradation. While some experiments produced inconclusive results, others gave direct evidence that a mechanical effect, the changing of gap thickness, could be directly related to the spectral shifts. Experiments such as the wedged cell and the CPR-730 clamp tests, which compressed the device, suggest that gap compression correlates to a blue shift. The disassembly of CPR-443 points to gap expansion as a possible cause of a red shift. The results of these experiments support the hypothesis that a changing gap thickness could be a cause of degradation. However, further investigation (Section 5) is required to fully understand the reason for LCP performance degradation.

5. Future Work

More research needs to be carried out to prove or disprove the hypothesis of a mechanically induced pitch distortion. Future work will include:

- Studying a correlation (if any) between λ_c and barometric pressure

  It is possible that barometric pressure could play a role in changing the gap thickness of the CPRs on a day to day basis. The central wavelengths of several existing CPRs will be monitored, and the central wavelengths will be compared to daily fluctuations in barometric pressure. Building CPRs in a controlled environment and with known gap thicknesses will also allow investigation of this possible correlation, by facilitating study on changes in gap thickness.

- Building new cells using old LC fluid

  A CPR would be disassembled and the LC fluid retrieved. That LC fluid would then be used to build a new CPR. The optical properties and λ_c of both the old and new CPRs could be analyzed and compared to determine if the LC fluid is degraded, or if its surroundings could be causing the λ_c shift. The LC fluid from CPR-443 could potentially be used for this type of experiment.

- Investigating the possibility of chemical degradation of the LC fluid

  Chemical degradation could be caused by numerous factors, such as degradation by flashlamp light, by laser light, or by reactions with the epoxy seal.

  Crystallization outside the clear aperture of several degraded devices was also observed. Properly functioning devices will be examined for the presence of crystals, and the cause of this crystallization will be researched.

- Utilizing the Exicor 450XT (Mueller Matrix) Polarimeter

  Due to the optical activity of the LC fluid, the polarimeter can be used to measure the UV optical rotation of the CPRs, and a technique utilizing this property will be developed for analyzing the thickness of the gap and LC fluid.

6. References

[1]: Chandrasekhar, S. 1993. Liquid Crystals. Cambridge Monographs on Physics. Bristol, England. Page 268. Note that the full equation is used in Appendix A.

[2]: Jacobs, Stephen D.; Smith, Douglas; Rigatti, Amy; Marshall, Ken; Puchebner, Birgit; and Lund, Lance. 1993. *Manufacturing Procedure, Liquid Crystal Optics, F-FJ-P-01 Revision A, OMEGA Upgrade*. pgs 56-59. Laboratory for Laser Energetics (January18th, 1993)

[3]: Kosc, Tanya Z. and Kelly, Jack. 6 March 2012. Laboratory for Laser Energetics Presentation: *In-Situ Liquid Crystal Polarizer Transmission Measurements: Test Fixture and Preliminary Data*.

[4]: Bailey, C. A.; Tondiglia V. T.; Natarajan, L. V.; Bricker, R.; Cui, Y.; Yang, D.K.; and Bunning T.J. *Surface limitations to the electro-mechanical tuning range of negative dielectric anisotropy cholesteric liquid crystals*. Journal of Applied Physics **111**, 063111 (2012)

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Appendix A

Ferguson's equation[1] gives the wavelength of maximum reflection (central wavelength, λ_c):

$$\lambda_c = nP\cos[\tfrac{1}{2}\, sin^{-1}(n^{-1}sin\varphi_i) + \tfrac{1}{2}sin^{-1}(n^{-1}sin\varphi_s)],$$

where P is the pitch length, φ_i is the angle of incidence, φ_s is the angle of scattering, and n is the mean refractive index of the LC.

Assuming that the incident light is normal to the pitch, we can let $\varphi_i = \varphi_s = 0$. Thus, $\sin(\varphi_i) = \sin(\varphi_s) = 0$, and we have $\lambda_c = nP$.

Appendix B

Equation 1 tells us that $\lambda_c = n \cdot p$. Thus, $p = \frac{\lambda_c}{n}$.

Assuming an average refractive index ~1.57, the theoretical pitch length for a λ_c of 1060 nm is

$$p = \frac{1060\, nm}{1.57} \approx 675\, nm.$$

The theoretical pitch length for a λ_c of 1090 nm (a 30 nm shift to the red) is

$$p = \frac{1090\, nm}{1.57} \approx 694\, nm.$$

This corresponds to a 694/675 = 1.028 or 2.8% increase in gap thickness.

A 2.8% increase in an original gap thickness of 20 microns is 20.564 microns, or a 564 nm change in gap thickness.

The theoretical pitch length for a λ_c of 1020 nm (a 40 nm shift to the blue) is

$$p = \frac{1020\ nm}{1.57} \approx 649\ nm.$$

This corresponds to a 649/675 = 0.9615 or 100% - 96.15% = 3.85% decrease.

A 3.85% decrease in an original gap thickness of 20 microns is 19.23 microns, or a 770 nm change in gap thickness.

Appendix C

One set of "old" CPRs was left in the Materials Lab for approximately 15 years. The age, LC fluid composition, substrate type, type and thickness of the spacers, epoxy composition, and coating of the CPRs are unknown. Their diameters are 100 mm. All of these CPRs are red shifted, and many have large areas of bubbles as well as LC fluid.

Table C1. Characterization of "old" CPRs

CPR Designation	**Appearance**	**Approx. λ_c (nm)**
CPR-132	No air bubbles	1095
CPR-443	No air bubbles. Currently partially disassembled.	1116
CPR-505	No air bubbles. Disassembled.	1056
CPR-693	No air bubbles.	1066
CPR-715	Approx. 50% air bubbles, 50% LC fluid.	1164
CPR-724	No air bubbles.	1172
CPR-730	No air bubbles	1108
CPR-732	Approx. 35% air bubbles, 65% LC fluid.	1140
CPR-733	Approx. 25% air bubbles, 75% LC fluid.	1120

The second set comprised “new” CPRs removed from the OMEGA system

Table C2. Characterization of “new” CPRs			
CPR designation	**Lowest λc (nm)**	**Highest λc (nm)**	**Appearance**
CPR-3	1065	1065	100-mm, no crystallization
CPR 4-1-16	1018	1025	135-mm, crystallization outside clear aperture
CPR 4-1-36	1027	1037	135-mm, crystallization outside clear aperture
CPR 4-1-106	1105	1111	135-mm, minimal crystallization outside clear aperture
CPR 4-1-138	1036	1039	135-mm, minimal crystallization outside clear aperture
CPR 4-1-139	1142	1151	135-mm, no crystallization
CPR 4-1-141	1099	1108	135-mm, minimal crystallization outside clear aperture
CPR 4-1-146	1025	1039	135-mm, crystallization outside clear aperture

Appendix D

The epoxy seal of CPR-505 was removed using a razor blade and pick. The hydrostatic pressure of the LC fluid prevented the substrates from being sheared apart, so the CPR was placed on a Thermolyne (Sybron) Nuova II hot plate. It was covered with a petrie dish in order to heat the CPR more uniformly.

The initial temperature of the hot plate was 22°C. The hot plate dial was set to 3 (approximately 50° C), and after about 15 minutes, air bubbles began forming about the circumference of the CPR and slowly moving towards the center. After another 10 minutes, the dial was turned to 4 (75°C – 84°C), and the temperature of the top substrate was measured to be 46°C.

Throughout the experiment, the air bubbles continued to redistribute and migrate toward the center of the CPR. By observing the interference fringes in the air bubbles, it was deduced that the CPR was bowing out at the center. After 45 minutes of heating, the substrates were sheared and successfully slid away from each other.

A Teflon spatula was used to scrape the LC fluid from each substrate into a small vial. The remaining LC was rinsed into a petri dish using hexane, and the hexane/LC fluid mixture was poured into another small vial. This vial was placed on a hot plate at ~24°C, and the hexane was evaporated off over the course of approximately 24 hours.