PCLC Flakes for OMEGA Laser Applications

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Abstract:

The OMEGA laser system uses over 300 liquid crystal (LC) optics to control the polarization of light. These optics are temperature sensitive and need thick, expensive glass substrates to contain the LC. Polymer liquid crystals (PLC) are much less temperature sensitive than low-molar-mass LC’s and do not require thick substrates, but they are extremely difficult to align over a large area. After breaking up well-aligned small-area films of PLC’s into particles called flakes, we are able to align them over large areas, potentially resulting in improved performance over the entire optic. This project studied the viability of using polymer cholesteric liquid crystal flakes for LC optics in the OMEGA laser system. This was achieved by experimenting with the concentration of flakes in a host fluid, the density and index of refraction of the host fluid, the size of the gap between the substrates, and the techniques used to assemble an optic.

1. Introduction

1.1 Liquid Crystals

Liquid crystal (LC) is a state of matter in between liquid and solid [1]. LC's flow like a liquid would, but they have more order to their molecular structure. Solids have both positional and orientational order, LC's have orientational order, and liquids have no molecular order. Some LC's have a rod-like molecular shape. The average direction in which the longest axis of the rod-like molecules is aligned is known as the director. LC's are anisotropic, meaning their properties change depending upon whether you observe them parallel or perpendicular to the director. One main property that changes is the LC's index of refraction. The difference in values in each direction for the index of refraction is called birefringence ($\triangle$n). Birefringence gives LC's some of their unique optical properties.

Substances that have an LC phase only stay in this phase over a specific temperature range. There are different types of LC's, and one especially interesting type is the cholesteric LC (CLC). This is one of the more complex mesophases, having a large amount of organization. As the layers of molecules lay over each other, the direction of the director twists with respect to the layer above it. This forms a helical structure in the LC (figure 1.1). This molecular

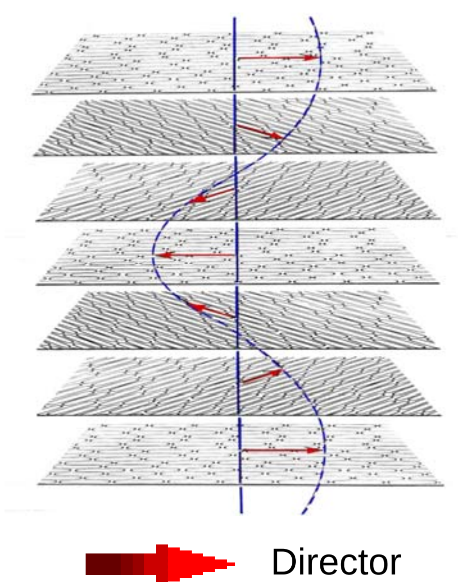


Figure 1.1- Shows the twist of the director through layers of the cholesteric LC molecules. Notice the helical structure. The pitch length refers to the distance for the director to complete a 360-degree rotation.

order gives the CLC its special property of selective reflection. The wavelength of electromagnetic radiation that a specific cholesteric LC will reflect depends on the average index of refraction of the LC, the angle of incidence of the light, and the pitch length of the CLC. The range of wavelengths reflected is dependent upon the angle of incidence, pitch length, and the LC's birefringence [2] (figure 1.2).

When the selected wavelength of unpolarized light enters a cholesteric LC, the component of the light that is circularly polarized in the same direction as the spiral of the LC structure will pass through unaffected. The other half of the light, now circularly polarized in the direction opposite that of the LC structure, will be reflected back and not pass through the LC.

$$\boxed{\lambda_r = \overline{n}_n P \cos\left\{\frac{1}{2}\left[\sin^{-1}\left(\frac{\sin\theta_i}{\overline{n}_{\text{ch}}}\right)+\sin^{-1}\left(\frac{\sin\theta_r}{\overline{n}_{\text{ch}}}\right)\right]\right\}}$$

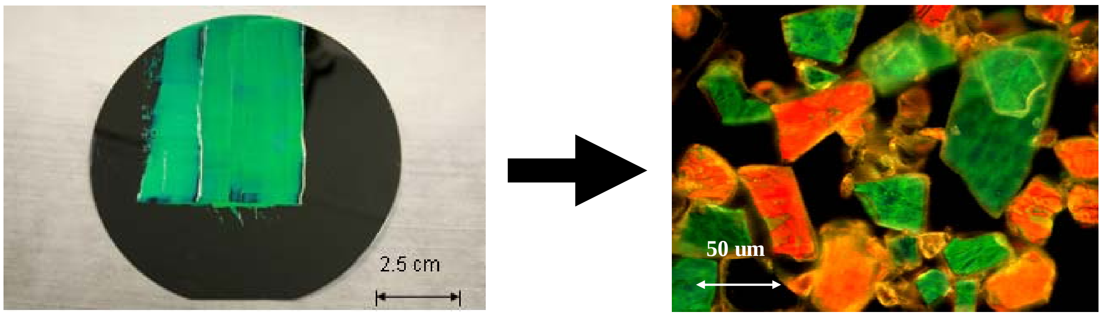
Figure 1.2- Formula for determining the wavelength of maximum reflection in a CLC.

P = Pitch Length.
θ_i = Incidence angle.
θ_r = Observation angle in air.
$\overline{n}_n$ = The average refractive index perpendicular to the helix.
λ_r = The wavelength at the center of the selective reflection band.
$\overline{n}_{\text{ch}}$ = The approximate average cholesteric refractive index.

1.2 Polymer Cholesteric Liquid Crystal Flakes

LC's are extremely temperature sensitive. Very small temperature changes will change their properties, or even cause a phase transition to a solid or an isotropic liquid. When liquid crystal molecules, or mesogens, are incorporated into a polymer chain, the resulting polymer LC is not nearly as temperature sensitive.

One disadvantage of polymer LC's (PLC's) is that they can only be well aligned over small areas because of their extremely viscous nature. Since a PLC optic would require a large, uniform, well-aligned area they would be difficult to utilize for this application. Small areas of well-aligned PLC can be frozen with liquid nitrogen, then broken into small solid flakes that preserve all of the important optical properties of LC's while remaining temperature insensitive (figure 1.3). These flakes vary in size, from tens to hundreds of microns. When cholesteric mesogens are used in this process it results in polymer cholesteric liquid crystal (PCLC) flakes [2].



Well-aligned, small-area PCLC film

Freeze-fractured PCLC flakes

Figure 1.3- PCLC flakes

1.3 Applications

One application in which PCLC flakes could be used is to replace the circular polarizer LC optics already in the OMEGA laser system. The PCLC flakes require a much less stable environment, allowing them be used in areas with less expensive climate control equipment (This is not an issue for Omega). The flakes devices can utilize much thinner substrates than low-molar-mass LC's, which require thick glass substrates to keep a uniform cell gap. The thicker the glass, the more distortion of the laser beam there is. Also, thinner substrates would be significantly cheaper.

A second application for the PCLC flakes would be to make a multi-wavelength filter [3]. By mixing multiple types of PCLC flakes in the same cell it is theoretically possible to make an optic that would selectively reflect multiple wavelengths. This would be useful for diagnostics purposes, such as for frequency tripling. An optic that reflects infrared and green light but transmits ultraviolet light could be used to reflect the infrared and green light onto a sensor to measure how much of each unwanted wavelength is present after frequency tripling. This would allow the efficiency of the frequency-tripling crystals to be determined.

2. Experiment

2.1 Host Fluid

When the PCLC flakes are put into a cell they are suspended in a host fluid. There are several highly desirable traits for this fluid to have. First, it must have the same index of refraction as the PCLC flakes in use. Light is scattered at each interface between materials with different indexes of refraction (figure 2.1). When trying to build

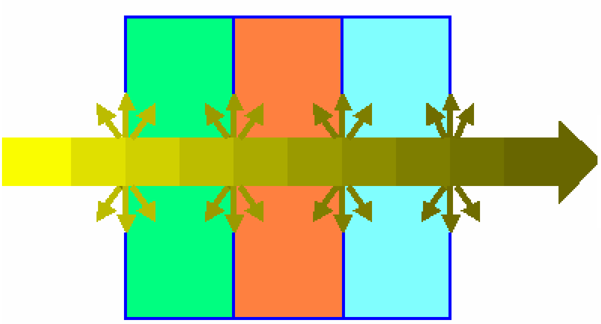


Figure 2.1- At each surface with a different index of refraction, light is scattered

a high-quality optic, scatter is a major problem. If light is scattered it is not transmitted down the laser path, decreasing the efficiency of the entire system. In order to get a fluid with the desired index of refraction and still be able to meet the other criteria necessary for a host fluid, different liquids were mixed together. This allowed a mixture to be produced with exactly the desired traits.

Second, the fluid should have the same density as the flakes. This will keep the flakes suspended in solution, allowing an optic to be turned at any angle while still giving consistent results. Having matching densities is ideal, but not absolutely necessary. If the flakes have a different density than the fluid, they should align against one of the substrates. This may be a useful technique for encouraging alignment. The drawback to it would be that the optic could only be used in a perfectly horizontal position or with a thinner than optimal cell gap to keep the flakes from moving. I made several mixtures of fluids, some that had densities that matched the flakes and some that had different densities, to explore the relationship between mismatched densities and flake alignment.

Since the host fluid is a mixture of its component liquids, it is essential that the components are miscible with each other. To determine this, I conducted a series of miscibility tests using the liquids I planned to use in my host fluids. I took mixtures of liquids, put them into small bottles, and shook them. Only fluids that remained clear when shaken were miscible and could be used as host fluids for the PCLC flakes.

The third important requirement for the host fluid is that it must be chemically compatible with the flakes. Some fluids will dissolve the flakes, rendering them useless. In order to test for chemical compatibility, I took samples of all the fluid mixtures that met the previous standards and added a small amount of flakes to the fluids. The fluids that were chemically compatible shimmered with the light being reflected off the flakes. Since the angle of incidence partially determines the wavelength of light reflected, the bottle of fluid and flakes shimmered with many colors and was very bright. When fluids were not chemically compatible it was immediately obvious. When flakes were added the fluid turned the dark, murky brown color of muddy water. The flakes dissolved into the fluid and discolored it. Only solutions that shimmered with many colors could be used as possible host fluids.

2.2 Concentration of PCLC Flakes

An LC optic will tend to reflect all the light of one handedness (50% of the total for unpolarized incident light) at its selected wavelength while allowing all other wavelengths of light to pass through. In order for PCLC flakes to achieve this goal, there must be a large number of flakes that are all well-aligned in a cell. They must uniformly cover the entire aperture of the optic to produce a uniform beam of reflected light. One of the most important aspects of determining the viability of a PCLC circular polarizer is getting enough flakes into the cell gap. No matter how well aligned they are, if there are not enough flakes the device will not be efficient enough to be utilized. In order to test how many flakes an optic can hold effectively under a given set of parameters, I built small test cells to simulate the properties of an optic. I used microscope slides as substrates, and cut them to about 2/3 of their original length to make the area in which I had to align the flakes smaller. By mixing glass microspheres into UV epoxy, and then adhering two slides together by putting dabs of epoxy at each of the four corners, I created a test cell that had a uniform gap, the size of which depended upon the microspheres used (figure 2.2). By holding every variable the same except for flake

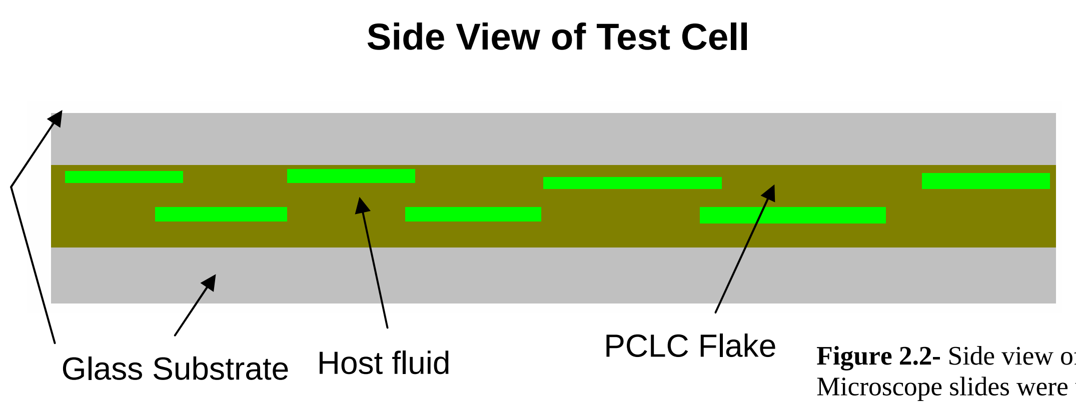


Figure 2.2- Side view of a test cell. Microscope slides were used as the glass substrate.

concentration in the host fluid, I was able to make a series of test cells that showed the relationship between concentration and maximum reflectance.

2.3 Alignment Techniques

The other important factor for determining the viability of a PCLC flake device is how well the flakes can be aligned. If they are not all lined up in the same plane, the flakes will target different wavelengths of light as the angle of incidence of the light affects the wavelength of light that will be reflected. Several different methods to force the PCLC flakes to align were tried. One was changing the characteristics of the host fluid, namely its density. Another was using different methods to construct the test cell. These methods should be able to be adapted to the construction of a full-size optic. By keeping all the variables the same except for the alignment technique I was testing, I developed cells to be used as controls and cells to experiment with alignment techniques. By comparing these cells against one another, I was able to tell whether the technique helped in flake alignment.

2.4 Cell Gap

The cell gap is a significant variable, but not directly related to the efficiency of the optic. The amount of flakes that can be fit into an optic (concentration) and how well they are aligned will ultimately determine whether PCLC-flake circular polarizers are feasible. The gap between the two substrates is important because it directly affects both the concentration limits of flakes in the host fluid and how well they are able to be aligned. In order to determine the best possible size for the cell gap, I first had to find the best way to align the flakes and what the optimum concentration of flakes was. Then, using these parameters to build several test cells that were identical except for the gap between the substrates, I was able to compare the results from these test cells and determine the best possible size for the cell gap. The results from these experiments are given in section **3.4**.

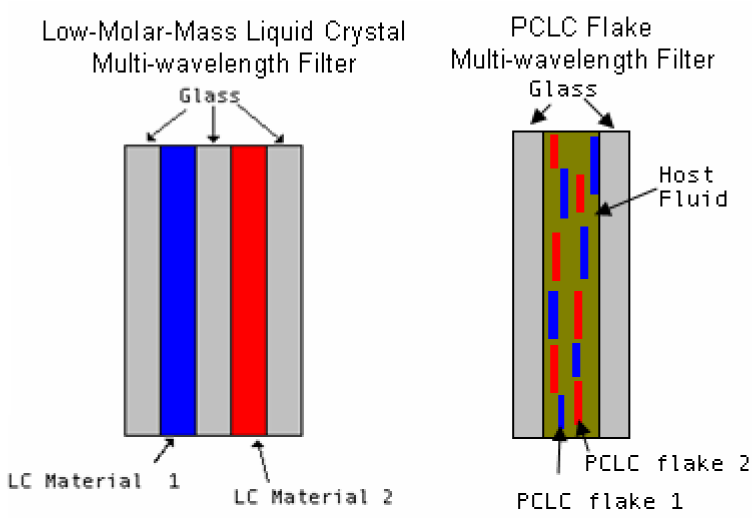
2.5 Multi-wavelength Filters

Multi-wavelength filters are an extension of circular polarizers. When constructed using low-molar-mass LC's, multi-wavelength filters are circular polarizers layered together into one larger optic. Each individual layer of LC reflects one wavelength of light, and the multiple layers of different LC result in a multi-wavelength circular polarizer. Because of the need to have layers of glass separating each type of liquid crystal, there is a large amount of scatter and specular reflection in these optics.

PCLC flakes can theoretically make more efficient multi-wavelength filters. Since different types of flakes that would reflect different wavelengths of light do not react with each other, it should be possible to put multiple types of flakes into the same cell gap. This type of optic would require much less glass than comparable low-molar-mass LC filters, meaning there would be less unwanted reflections and less scatter (figure 2.3).

All the challenges of making a PCLC flake circular polarizer would be magnified in a multi-wavelength filter. There would need to be a great amount of several different types of flakes, and they would all have to be well-aligned. In essence, if a device is built with two reflective wavelengths, the requirements in concentration and alignment for a circular polarizer must be met twice over. The main goal of this project is to attempt to prove a single-wavelength polarizer viable. Testing on multi-wavelength filters shows that the scope of this project can be expanded upon.

Figure 2.3- A low-molar-mass liquid crystal multi-wavelength filter compared to a PCLC flake multi-wavelength filter. The PCLC flake filter uses much less glass, which is expensive and increases scatter.



3. Results

3.1 Host Fluid

Through the process described in section **2.1** I found a host fluid that matched all the desired criteria. The calculations for determining the index of refraction and density of a fluid made from two components are based upon a linear relationship to the percent composition of each liquid. In the search for the right host fluid I conducted tests using mixtures that included Bromonaphthaline, PDM-7050, propylene carbonate, DMS-T12, Mistie, and DMS-T31. I found the ideal fluid in the form of a mixture of 10.7% propylene carbonate and 89.3% PDM-7050. It had the same index of refraction, 1.57, as the flakes I was using, was chemically compatible with the flakes, and the components were miscible with each other. The density of the fluid was nearly ideal, too. The density of the flakes I used was ~1.10 g/cm^3, and the fluid's density was calculated to be 1.101g/cm^3.

3.2 Alignment Techniques

I tried several different techniques to align the PCLC flakes. The first was manipulating the host fluid. By having a fluid with a density that was different from that of the flakes, the flakes were forced to align against one of the substrates. An interesting finding was that when the density of the fluid was greater than that of the flakes, forcing them to rise to the surface, the cell reflected a higher percentage of light at its targeted wavelength, yet when the density of the fluid was less than that of the flakes, allowing them to sink to the bottom, the reflection at the targeted wavelength actually decreased. By using a host fluid that had a density 0.1g/cm^3 greater than that of the flakes, maximum

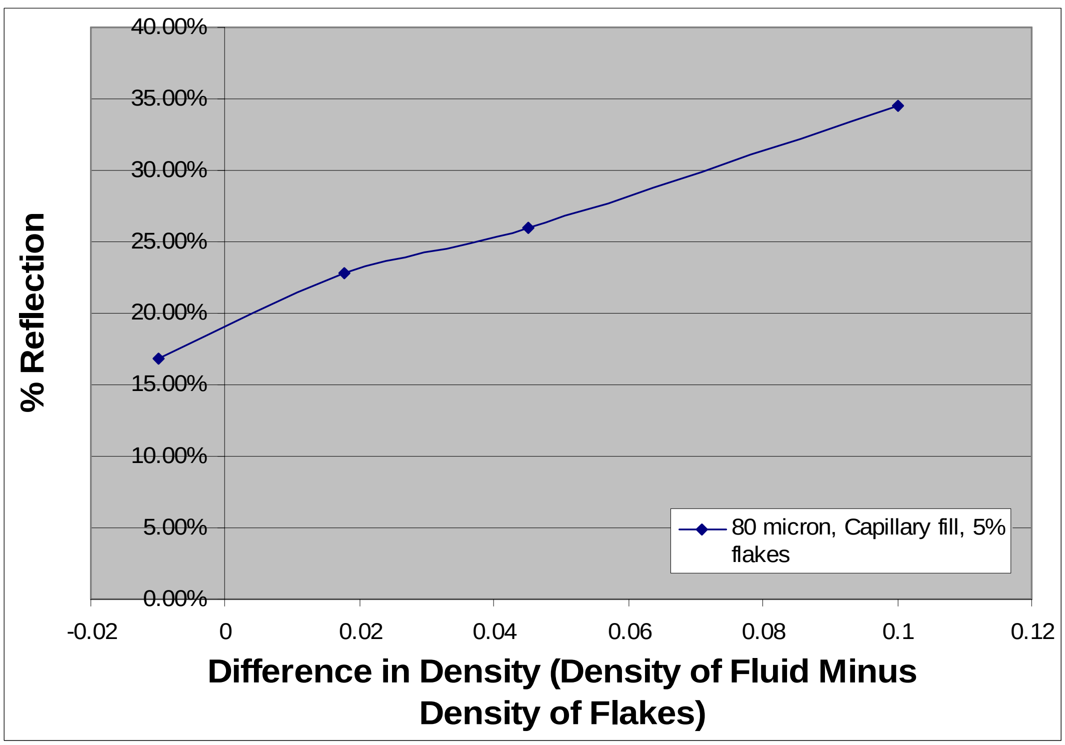


Figure 3.1- Dependence of the reflectivity on the density of the host fluid relative to the PCLC flakes. All factors except for the density of the host fluid were held constant. The relationship is nearly linear.

reflection was increased substantially from 22.8% to 34.6% (figure 3.1). The 34.6% reflectivity was the highest observed in any of the tests. This shows that a density difference could be an extremely effective alignment technique if the optic it was applied in could stay perfectly horizontal throughout its entire operating life. Since this is not the case in the OMEGA laser system at the moment, I had to find another method for aligning the PCLC flakes in my test cells.

The other method I used in an attempt to improve alignment was to alter the way the test cells were built. Traditionally, test cells were constructed by adhering the two substrates together and then filling the gap with the PCLC flakes in their host fluid by utilizing capillary action. I tried two new methods of construction of the cells. The first method was to put the PCLC flakes in the host fluid on the bottom substrate, and then put the top substrate on and shear the substrates against each other moving in the longer direction of the microscope slides (figure 3.2). This improved upon the results from the

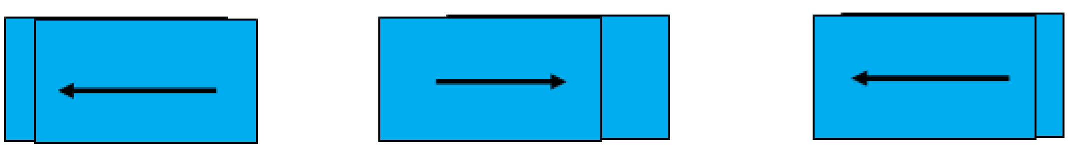


Figure 3.2- The shear method. Arrows show which way the top substrate slides over the bottom substrate.

capillary fill method used previously, increasing reflection from the previous maximum of 22.8% to 28.5%. The second new construction method was the "twisting shear" method. It also started with putting the PCLC material on the bottom substrate, but then the top substrate was twisted back and forth across the bottom substrate (figure 3.3). This improved reflection even more than the first shearing method I experimented with (figure 3.4), reaching a maximum reflection of 30.18% at a concentration of 7.5% flakes by weight.

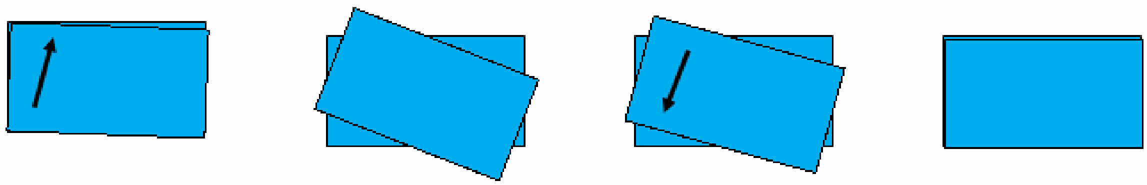


Figure 3.3- The "twisting shear" method. The arrows show which way the top substrate slides over the bottom substrate. This is the most effective method of aligning the flakes.

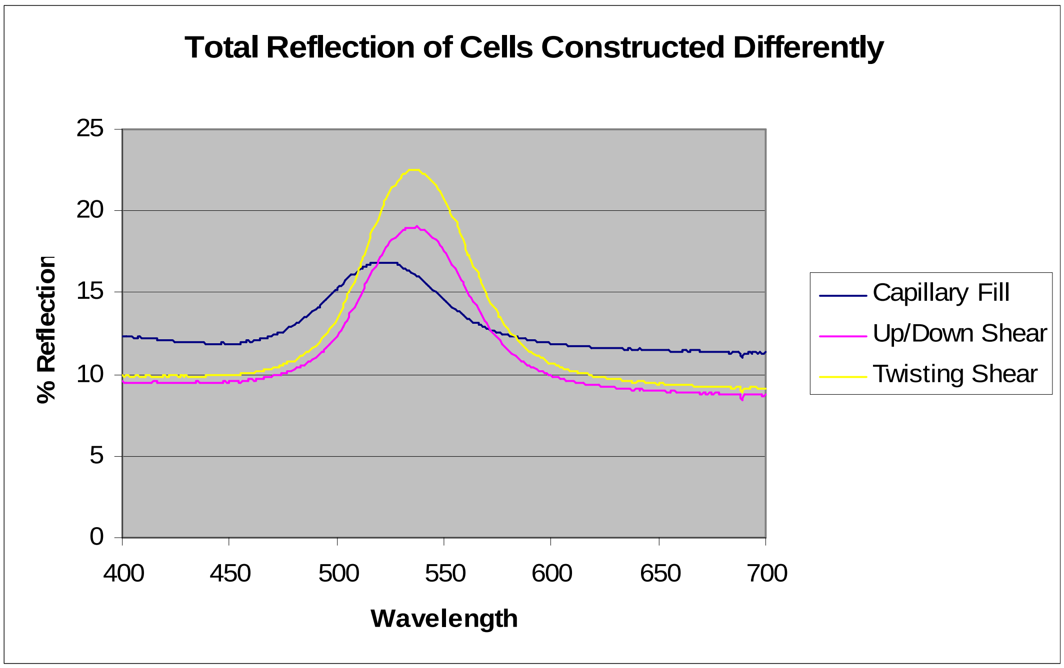


Figure 3.4- A graph showing the total reflection of cells constructed three different ways. All cells were built with an 80 um cell gap using 5% flakes by weight. The only differences between the cells are the methods used for construction. The twisting shear technique gives the highest reflection of any construction method. The differences in wavelength of maximum reflection and wavelength ranges are brought about by flakes that are not perfectly aligned, altering the angle of incidence of the light on the flakes. Note: These reflectivities are not the maximum values for the construction methods. The cells were made using less than optimal flake concentration.

3.3 PCLC Flake Concentration

From previous work done on PCLC flake concentration in an optic it was concluded that the optimum concentration of flakes in percent by mass was 5% [3]. This work used the typical construction method for test cells and relied upon capillary action to fill the cell gap. With my new alignment technique of "twisting shear", I was able to bolster the optimum concentration to 7.5% flakes by weight, an increase of 50% over the previous concentration achieved (figure 3.5).

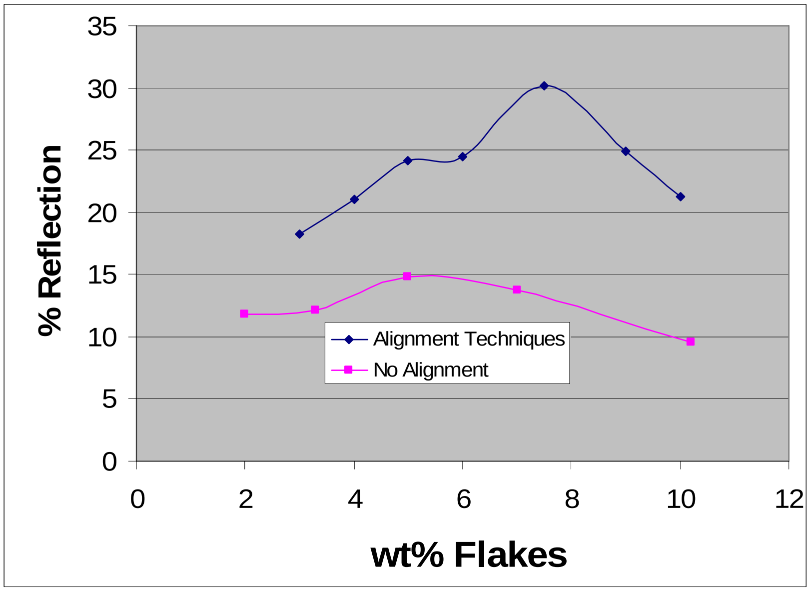


Figure 3.5- A graph showing the reflectivity of cells vs. the concentration of flakes in percent by weight for cells in this study (aligned) and for previously made cells (not aligned). The alignment techniques drastically increased the amount of flakes that can be effectively inserted into a device, thereby increasing reflectivity. The optimum amount of flakes was increased from 5% by weight to 7.5% by weight.

3.4 Cell Gap

In initial experiments, when finding out the optimized concentration of PCLC flakes in a test cell filled by capillary action, there was no variation in cell gap [3] [4]. It was not known how changing the cell gap would affect the concentration and alignment of flakes. Through my tests, regardless of what construction method was used, an 80 um cell gap yielded the highest consistent reflectivities. The reflectivity of my test cells drastically decreased when the cell gap decreased. The maximum reflection for a test cell made with an 80 um cell gap was 30.18%, while the maximum for a 40 um cell gap was only 22.49%. When the cell gap is increased to over 80 um, the reflectivity of the cell varied randomly. This shows that the flakes have too much room to float around in and are constantly changing their alignment. It seems that an 80 um cell gap allows for the greatest number of flakes in the cell while keeping them well-aligned, yielding the highest consistent reflectivities.

3.5 Multi-wavelength Filters

I made a two-color filter with flakes reflecting in the red and flakes reflecting in the blue range. The reflectance measured by a spectrophotometer clearly showed two peaks, one in the red wavelength range and one in the blue range (figure 3.6). This proved that the concept of a multi-wavelength PCLC flake filter is viable. Using 5% by mass of each kind of flake for a total of 10% flakes, I was able to reach a maximum of 22% reflection at each peak, while still letting the same amount of untargeted light transmit through as in a single color PCLC circular polarizer. This is a very promising area for future study.

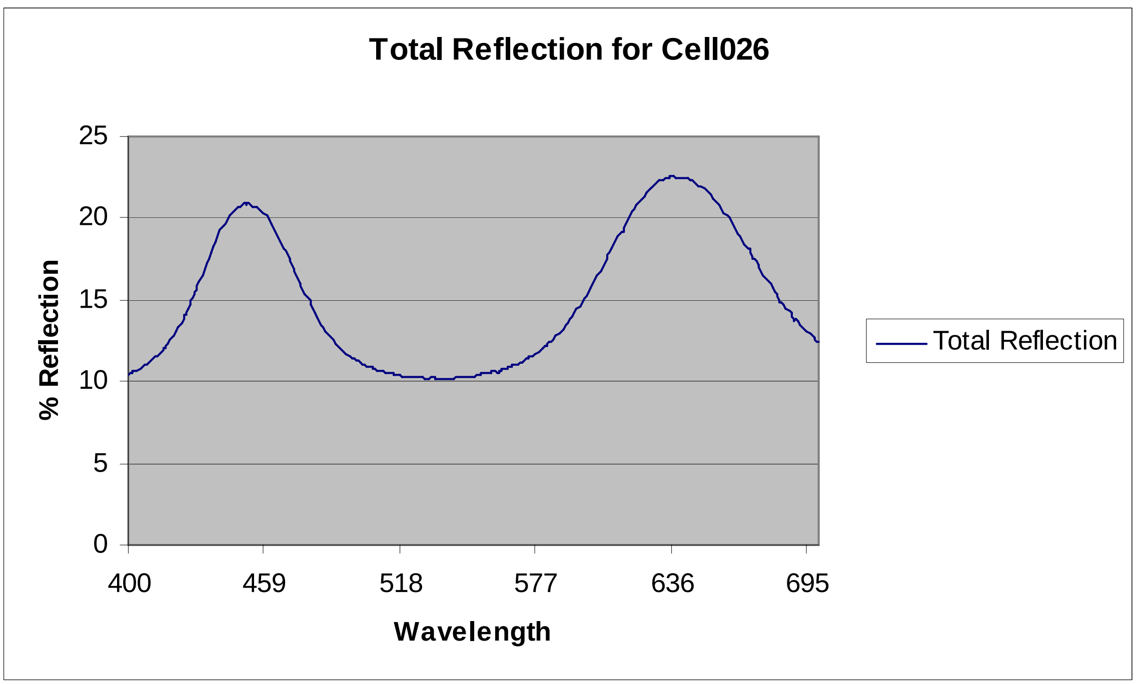


Figure 3.6- A graph showing reflectivity vs. wavelength in a two-color PCLC multi-wavelength filter. One PCLC flake type reflects light in the blue range and the other PCLC flake type reflects light in the red range. Notice the two distinct peaks in the graph. Each one represents the targeted wavelengths of one kind of PCLC flakes. Reflection went down to normal levels between the peaks, indicating that the two flake types did not interfere with each other.

4. Conclusion/ Future Work

4.1 PCLC Circular Polarizers

The goal of this work was to determine the viability of a PCLC flake circular polarizer. Even though significant advances were made in this work, it is likely that more improvement will be made in the future. Previously, the maximum reflectivity achieved with a PCLC-flake circular polarizer was 14.8%. Through the use of new construction techniques that facilitated flake alignment and increased the maximum amount of flakes that can be used in a cell, the reflectivity was increased to 30.18%, doubling the effectiveness of these cells (figure 4.1). There is still room for improving the reflective

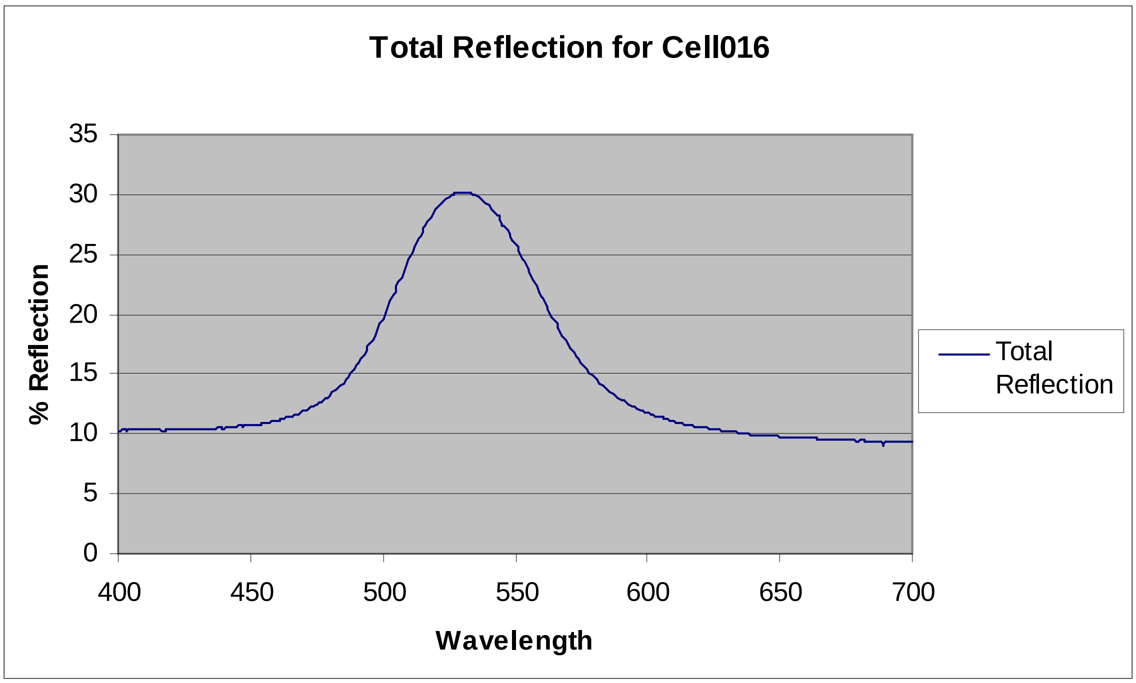


Figure 4.1- The graph of reflectivity vs. wavelength for a cell constructed using the twisting shear method, an 80 um cell gap, and an ideal host fluid with 7.5% flakes by weight suspended in it. The maximum reflectivity I was able to achieve was 30.18%. This is the highest reflectivity in this work that uses a density-matched fluid.

characteristics of these devices, such as using a UV curable polymer to lock well-aligned flakes into place while using only one substrate or wetting the base substrate, sprinkling hydrophobic flakes onto the wet glass, then allowing the water to dry, leaving the flakes that had been on the surface of the water to lay flat on the substrate. With more experimentation and new methods for the alignment of the PCLC flakes, it could well be possible to use PCLC flake circular polarizers in the OMEGA laser system.

4.2 Multi-wavelength Filters

Multi-wavelength PCLC flake filters were tested to determine whether the theory behind them can is valid. The concept was demonstrated through reflectivity measurements taken on a test cell using multiple PCLC flakes that targeted different wavelengths. The graph of reflectivity vs. wavelength plotted for a two-colored circular polarizer clearly showed two distinct peaks characteristic of the selective reflection properties of the two types of PCLC flakes. The two types of flakes in the cell did not interfere with each other. Further work on this type of optic will be carried out to evaluate its potential for use in diagnostics in the OMEGA laser system because of the benefits that can be realized through this design.

5. Acknowledgments

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