Next Generation Liquid Crystal Mixtures for OMEGA Circular Polarizer/Isolator Devices

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Abstract

Liquid crystal (LC) devices have been in use in the OMEGA laser system for over 30 years both to generate circularly polarized light in order to optimize power output and to prevent against back-reflections that could damage the laser system. New LC mixtures for use in OMEGA LC circular polarizers (LCP's) must be developed because one of the components of the current LC formulation (ZLI-1167) is no longer available commercially. A survey of commercially available LC host materials and chiral dopants was conducted, and the most viable candidates were selected and used in the preparation of test mixtures for evaluation of key device properties such as selective reflection peak width, peak wavelength tunability, and laser damage threshold. Of the three LC mixture formulations that were developed and characterized, a mixture containing 11.65% of R811 chiral dopant in the nematic host MLC 6601 showed the best thermodynamic stability over time and possessed optical properties similar to those of the LC mixtures currently in use in OMEGA LCP devices, while also achieving significantly higher laser damage thresholds.

1. Introduction

In the OMEGA laser system, liquid crystal (LC) devices are used to circularly polarize near-IR light at 1054 nm in order to optimize power output as well as isolate one handedness of light, to preventing damage to laser system optics. These devices help to optimize power output when compared to linear polarizers based on the fact that all laser glass fabricated in large apertures will have minor imperfections in optical homogeneity due to stresses produced by the glass fabrication process. Consequently, linearly polarized light passing through this medium will be converted to elliptically polarized light, which results in a loss of energy of up to 30%

[1]. Circularly polarized light passing through the same medium will be unaffected by these imperfections, which allows the laser system to operate at nearly 100% efficiency. [1]

Unpolarized incident light contains two different circularly polarized components [right-handed (RH) and left-handed (LH)]; LC circular polarizers (LCP's) isolate one handedness of light from the other by allowing one handedness of light to pass through the device, while the other handedness is reflected or scattered. For LC materials, this "selective reflection" effect occurs for specific wavelengths that satisfy the equation $\lambda_{max} = \boldsymbol{n}p$, where λ_{max} is the selective reflection peak wavelength, $\boldsymbol{n}$ is the average refractive index of the mixture and p is the pitch length, which is dependent on the chiral dopant concentration, its helical twisting power, and the mixture temperature. [1] Liquid crystal polarizers are preferable to solid-state devices made from conventional optical materials such as mica or quartz due to attributes such as broad angular tolerance, better scalability to large apertures, high laser damage resistance, and substantially lower cost. An additional feature of LCP devices is their ability to prevent laser induced damage from circularly polarized light reflecting back into the laser system, as incident light undergoes a change in handedness when reflected back off the target (Fig. 1).

Liquid crystal mixtures suitable for use in LCP devices are formulated by introducing a chiral dopant (a compound that cannot be superimposed on its mirror image due to inherent asymmetry in its molecular structure) into a nematic LC host composed of a compound or mixture of compounds that are rod-like in shape and all point in roughly the same direction, represented by a vector called the director. [1] This chiral dopant imparts its twist sense to the host material, creating a chiral nematic LC mixture with a helical structure (Fig. 2).

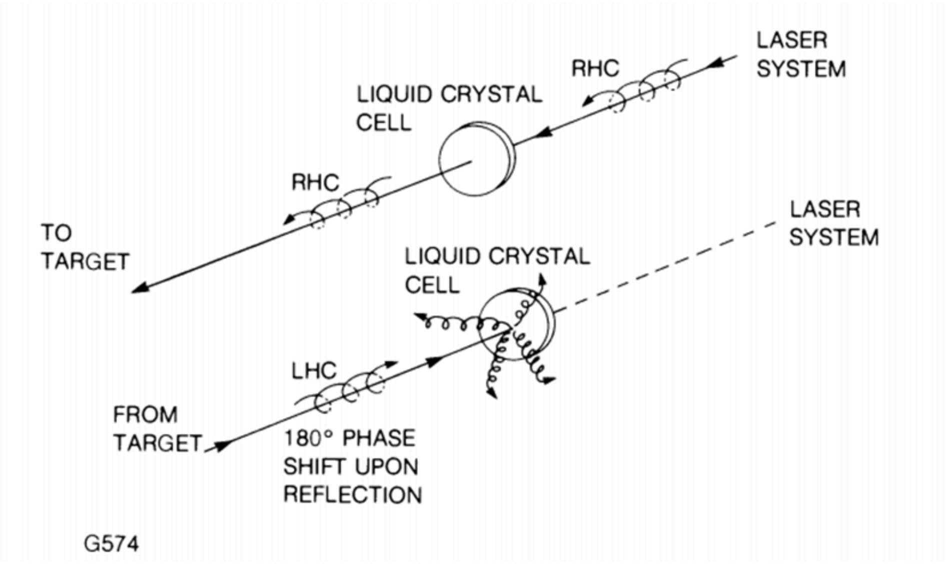


Fig. 1: Illustration of the use of LC polarizers to protect the laser system from back-reflected light. RHC and LHC denote the two polarizations of light.

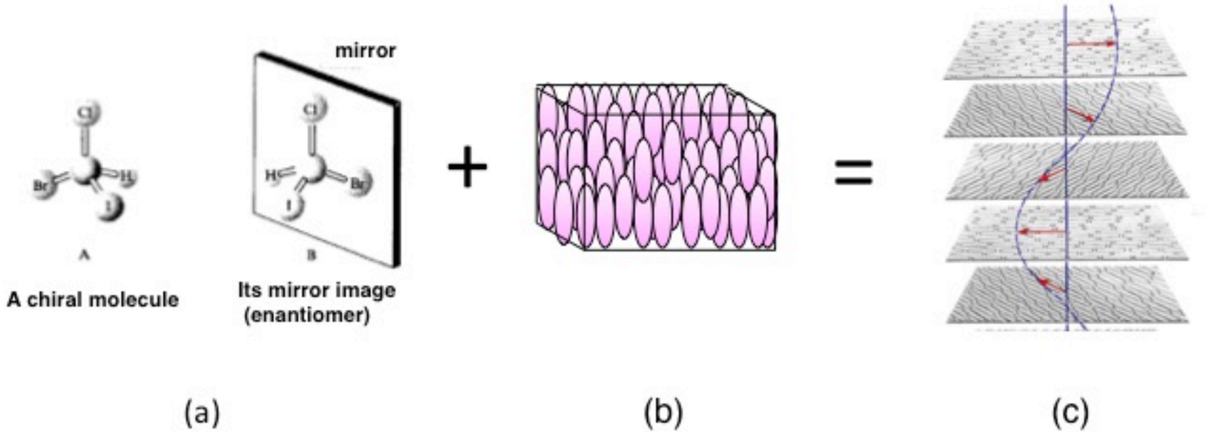
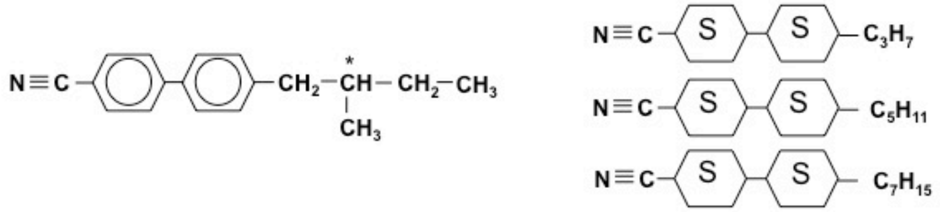


Fig. 2: Visual representation of the process of forming a chiral nematic LC mixture. The chiral dopant (a) is added to the nematic host (b) forming the chiral nematic helical structure (c) that has the ability to selectively reflect one handedness of circularly polarized light.

Existing RHLCP’s in OMEGA employ a mixture of commercial LC materials from EM Industries, Inc. (19.44% CB15 in ZLI-1167, shown in Fig. 3). [1] Because the ZLI-1167 nematic host is no longer commercially available, a new LC mixture must be developed in order to replace any RHLCP’s that have been damaged in the laser system.



CB-15 **ZLI-1167**

Fig. 3: The molecular structures of the chiral dopant (CB-15) and nematic host (ZLI-1167) used in the RHLCP devices currently deployed in OMEGA.

For this study, two commercially available nematic liquid crystal mixtures, MLC 6601 and MLC 2037, were evaluated as replacement hosts for ZLI-1167. These materials were chosen because of earlier work done at LLE that identified them as desirable candidates for LC device applications in high-peak-power UV lasers due to their high 351-nm laser damage thresholds [2]. Although no near-IR laser damage threshold data had been determined on these materials, it was expected that they would perform at least as well (and most likely better) in the near-IR due to their very low optical absorbance across the entire UV, visible, and near-IR regions. Two classes of commercially available chiral dopants were also evaluated; a right-handed cyanobiphenyl material (CB-15) and both right-handed and left-handed versions of a phenyl benzoate ester (R-811 and S-811, respectively), shown in Fig 4 below.

S-811 **R-811**

Fig. 4: The molecular structures of the phenylbenzoate ester chiral dopants R-811 and S-811.

2. Experimental

2.1 LC mixture preparation

Initial mixture compositions for testing were determined from previous compatibility studies of the chiral dopants CB-15 and S-811 in the nematic hosts MLC 2043 and MLC-6601 [3]. Preliminary experiments were conducted using the left-handed chiral dopant S-811 rather than right-handed R-811 due to the low cost and ready availability of the former and the high cost of the latter. This substitution is reasonable because the two optical isomers are essentially identical in every respect except their helical twist sense. A concentration of ~20% wt/wt of the chiral dopant in each nematic host was used as a starting point for the testing. The LC mixtures were prepared by combining the chiral dopant and nematic host in a clean beaker and heating the mixture on a hotplate with stirring until the LC material reached its isotropic phase and became completely transparent. The mixture was stirred and heated for 60 min in the isotropic phase to ensure it was completely homogeneous.

2.2 LC mixture optimization and test device assembly

Test samples were prepared by placing a small amount of the LC mixture on microscope slides that had been cleaned in a Class 10,000 clean room in the Materials Laboratory using an aqueous cleaning process that consisted of the following steps: (1) scrubbing with 0.05-µm MasterPrep® polishing suspension using a synthetic wiper, followed by rinsing with 2 megohm-cm deionized (DI) water and 18.5 megohm-cm DI water; (2) ultrasonic cleaning at 69°C in a solution of Extran detergent for 1 hr.; and (3) final rinsing with 18.5 megohm-cm DI water, followed by blow-drying with nitrogen to remove any visible moisture. Final drying was accomplished on a hotplate at 130°C for 60 minutes.

The selective reflection peak wavelength (λ_{max}) of the test mixtures was determined using a Perkin Elmer Lambda 900 UV-VIS NIR spectrophotometer. The concentration of the chiral dopant was adjusted, new test samples were prepared on microscope slides, and the samples re-tested on the Lambda 900 until the λ_{max} of the mixture was ~1060 nm. Once this λ_{max} value was achieved, the mixtures were used to construct LCP devices in the clean room area using two 25 mm dia. glass substrates that had been subjected to the same cleaning process as the microscope slides. One cleaned substrate was placed on a hotplate in the Class 100 clean hood housed within the clean room; to control the cell gap, a small amount of LC material was mixed with 14 µm microspheres in the LC isotropic phase and droplets of the mixture were placed at 120° intervals around the edge of the substrate. The second substrate was lowered onto the first substrate and the substrates were offset to produce a small ledge on which to deposit the LC mixture for filling the device. After heating the empty device to 90°C, a small amount of LC was dispensed onto the ledge using a syringe equipped with a submicron particle filter and the LC fluid was allowed to flow into the cell gap by capillary action. The hot plate was set to cool at a rate of 10°C /hr; once the device had cooled to 20°C, the substrates were bonded together using three small pieces of glass ("tacks") positioned around the outside of the device at 120° using an epoxy adhesive. The devices were then ready for determination of the λ_{max} using the Lambda 900 spectrophotometer.

3. Results and Discussion

Figures 5, 6, and 7 compare selective reflection scans for mixtures of the chiral dopants CB15 in MLC 6601 and MLC 2037 (Figs 5 and 6) and for S-811 in MLC 6601 (Fig 7) as functions of sample aging. The λ_{max} for both MLC 2037 and MLC 6601 containing 20% wt/wt of

CB15 drifted toward longer wavelengths as the samples aged over 7-9 months, while the λ_{max} of the mixture containing S-811 in MLC6601 remained essentially unchanged after 5 months.

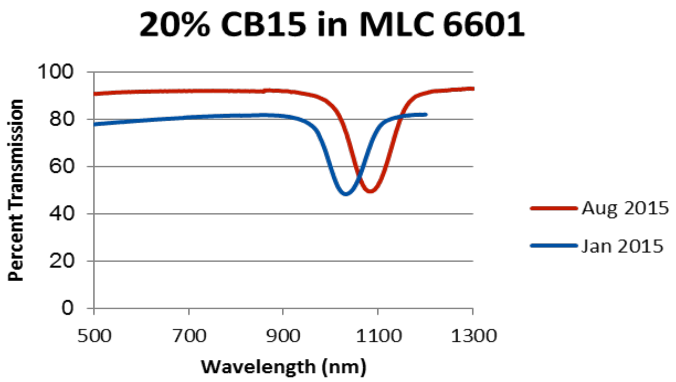


Fig. 5: Plot of the transmission of CB15 in MLC 6601 over time. The peak wavelength changed by 52 nm after 7 months.

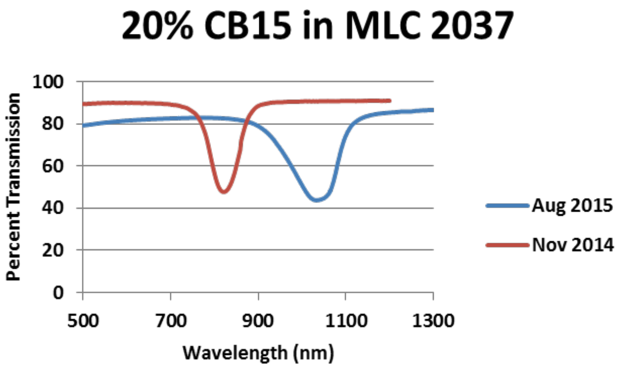


Fig. 6: Plot of the transmission of CB15 in MLC 2037. The peak wavelength changed by 212 nm in 9 months.

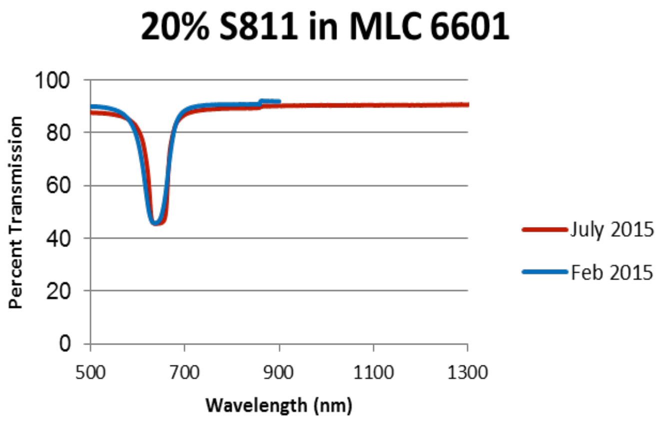


Fig. 7: Plot of the transmission of S811 in MLC 6601 over time. The peak wavelength changed by 4 nm in five months.

The cause for the drift in λ_{max} for the samples containing CB-15 is not well understood, but may possibly be due to micro-phase separation of the chiral component from the host material. No such shift in λ_{max} is observed with S-811 in MLC 6601; this chiral compound is significantly less polar than CB-15, which might reduce micro-phase separation. Further investigations centered on the S-811/MLC 6601 due to its good long-term stability.

The λ_{max} for the S-811/MLC 6601 mixture was fine-tuned to the operating wavelength of OMEGA by preparing a series of >15 mixtures with varying concentrations of S-811 in MLC 6601 and constructing a plot of λ_{max} vs dopant concentration using selective reflection data obtained from these mixtures (Fig. 8). [4]

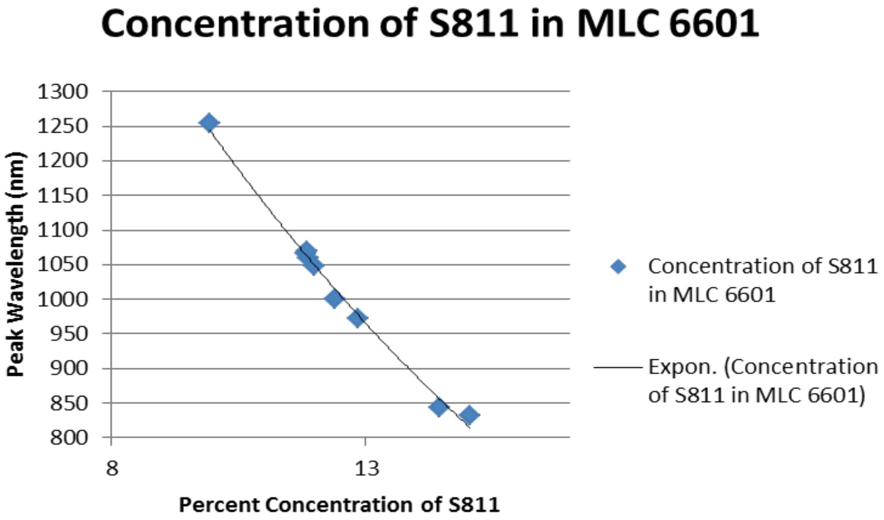


Fig 8: Scatter plot of the transmission peak of S811 in MLC 6601 vs. concentration. The data illustrates the wavelength tunability of the mixture. The line is an exponential fit to the data.

Because the chemical structure and physical properties of S-811 and R-811 are essentially identical except for their twist sense, the curve in Fig. 8 for S-811 in MLC 6601 was used to determine the required concentration of R811 in the same host to generate a λ_{max} at 1060 nm. Preparation of the R811/MLC 6601 mixture at this chiral component concentration revealed that the helical twisting power (HTP) of R811 in MLC 6601 was slightly greater than its left-handed analog, which required a slight reduction in R811 concentration to obtain the correct λ_{max}. Figure 9 shows selective reflection curves for concentrations of R811 in MLC 6601 ranging from 11.65% to 11.85%.; the 11.65% concentration displayed a λ_{max} of 1065 nm. Test LCP devices prepared with this composition showed values of λ_{max} that were was consistently 5-7 nm lower than those obtained for the same material measured between microscope slides, which necessitated additional fine tuning of mixture composition.

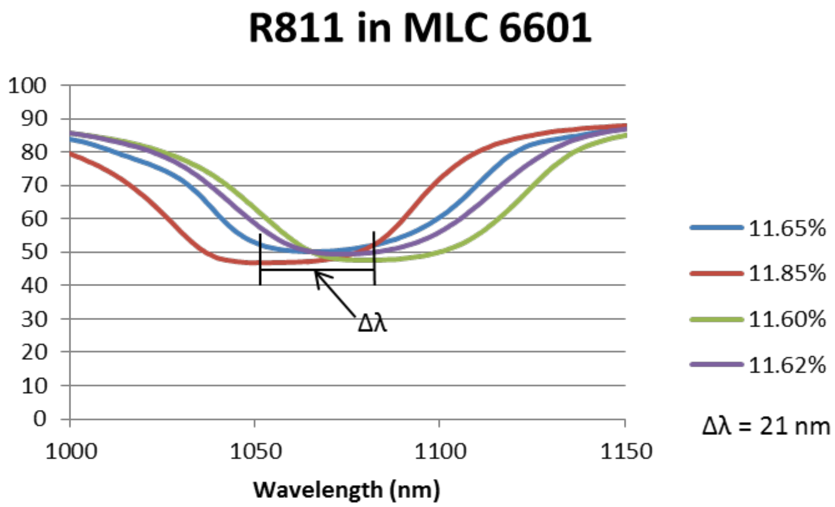


Fig. 9: Plot of the transmission of various concentrations of R811 in MLC 6601. An R811 concentration of 11.65% displayed a λ_{max} of 1065 nm, and was used to prepare test RHLCP devices.

. The optical properties of the R811/MLC6601 mixture were examined by both polarizing optical microscopy and reflectance spectroscopy to compare them to those of the CB15/ZLI 1167 mixture currently used in OMEGA RHLCP devices. Both materials showed nearly identical LC textures when viewed under crossed polarizers at 100x magnification using a Leitz Orthoplan Pol polarizing microscope in transmission. Reflectance data was also collected for the R811/MLC6601 device using the Lambda 900 spectrophotometer equipped with an integrating sphere capable of measuring total, diffuse, and specular reflection components. Un-polarized incident light was used for these measurements. The reflectance data showed that the majority of light reflected from the device is diffuse and not specular, indicating that most of the opposite handedness of the light scatters when it hits the LC mixture. Figure 10 shows selective reflection spectra for the R811/MLC6601 device compared to that for an RHLCP device from OMEGA (CPR 4-1-65); the difference in λ_{max} between the two mixtures is only ~1 nm, and the difference in peak widths is within 5 nm.

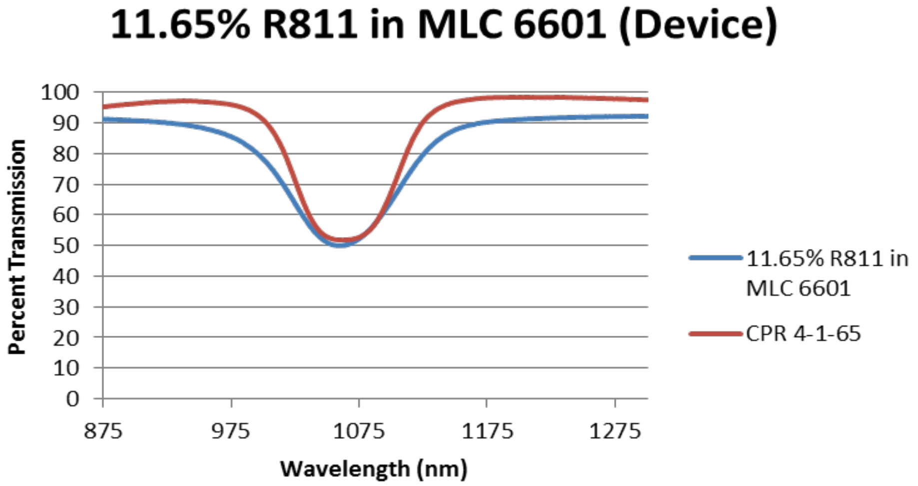
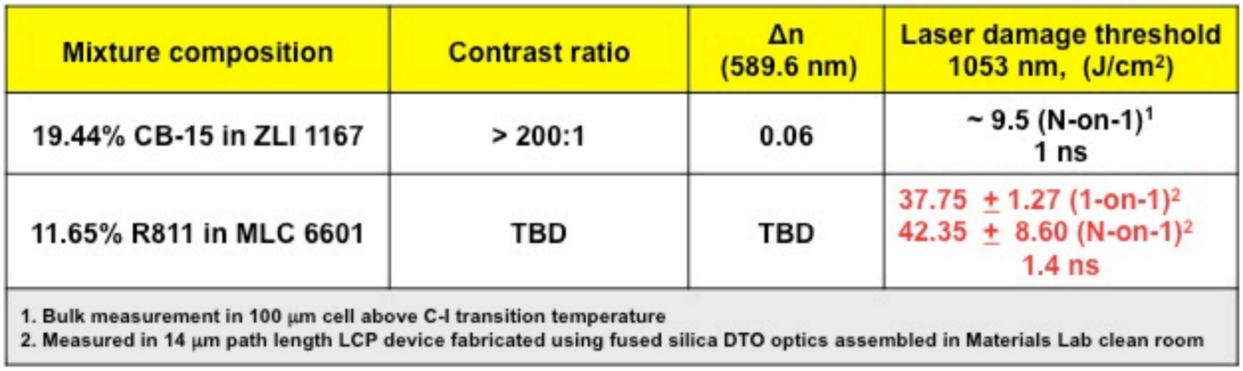


Fig. 10: Selective reflection spectra for 11.65% R811 in MLC 6601 and an OMEGA RHLCP device (CPR 4-1-65) containing 19.44% CB15 in ZLI-1167. The higher transmission of CPR 4-1-65 is due to both anti-reflection (AR) coatings on the external substrate surfaces and Nylon alignment coatings on the inner cell surfaces, neither of which are present in the R811/MLC6601 device.

Devices for laser damage testing were prepared in the same manner as described in Section 2.2, but instead using 60-mm-dia, 10-mm-thick fused silica damage test optics (DTO) substrates specifically intended for preparation of damage test samples. Damage testing was conducted at 1054 nm in the chiral nematic LC phase using both 1-on-1 and N-on-1 testing protocols with a 1.4 ns pulse width. In 1-on-1 testing, different sites are irradiated with a single shot, with the laser fluence increased by 15% at each location until damage is observed. In the N-on-1 testing mode, only a single site is irradiated with increasing fluence until damage is observed. Table 1 lists the damage threshold data collected on the R811/MLC 6601 mixture at 1053 nm, along with previously obtained laser damage data on the CB15/ZLI-1167 mixture currently used in OMEGA RHLCP devices. The very high laser damage thresholds for the new R811/MLC6601 mixture represent a significant improvement over the existing formulation.



Mixture composition	Contrast ratio	Δn (589.6 nm)	Laser damage threshold 1053 nm, (J/cm^2)
19.44% CB-15 in ZLI 1167	> 200:1	0.06	~ 9.5 (N-on-1)[1] 1 ns
11.65% R811 in MLC 6601	TBD	TBD	37.75 ± 1.27 (1-on-1)[2] 42.35 ± 8.60 (N-on-1)[2] 1.4 ns

1. Bulk measurement in 100 μm cell above C-I transition temperature
2. Measured in 14 μm path length LCP device fabricated using fused silica DTO optics assembled in Materials Lab clean room

Table 1: Laser damage threshold data for the new R811/MLC6601 mixture at 1053 nm. Similar data on the RHLCP mixture currently used in OMEGA RHLCP devices is included for reference. Some optical properties for the new mixture have yet to be determined.

4. Conclusions

After testing several possible replacements for the current LC mixture used in the RHLCP devices in the OMEGA laser system, a mixture of 11.65% R811 in MLC 6601 was determined to be the most viable of the over 15 different mixtures that were tested. This mixture shows good thermodynamic stability, optical properties similar to the current mixture, and a significantly improved laser damage threshold (37- 42 J/cm^2 at 1053 nm, 1.4 ns). The new materials combination also exhibits ease in selective reflection wavelength tunability and a reflection peak width essentially equivalent to the current mixture used in RHLCP devices. Further work that will be required before this material can be fully qualified for use in OMEGA includes a full characterization of all optical properties (e.g. contrast ratio transmission, scatter) in a full-aperture (100-200 mm) RHLCP device with nylon alignment coatings and deployment of this device for full-aperture testing on OMEGA.

5. Acknowledgements

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6. References

[1] K. L. Marshall, private communication.

[2] D. Saulnier, B. Taylor, K. L. Marshall, T. J. Kessler, and S. D. Jacobs, "Liquid Crystal Chiroptical Polarization Rotators for the Near-UV Region: Theory, Materials, and Device Applications," in *Liquid Crystals XVII,* edited by I. C. Khoo (SPIE, Bellingham, WA, 2013), Vol. 8828, Paper 882807.

[3] K. Smith, Optical Materials Laboratory, Undergraduate interim research report, Jan 15, 2016.

[4] T. Z. Kosc, A. Owens, A. L. Rigatti, S. D. Jacobs, and J. H. Kelly "Long-Term Performance of Liquid Crystal Optics on Large Fusion Lasers," presented at CLEO 2013, San Jose, CA, 9–14 June 2013.