

Laser-Induced–Damage Behavior of Novel Glassy Liquid Crystal Materials at 1 ns and Multiple Wavelengths

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Low-molar-mass (LMM) liquid crystal (LC) fluids have demonstrated a proven track record of high-peak-power performance on the OMEGA Nd:glass laser at LLE since 1985. Over 320 large-aperture LC circular polarizers (LCP's) and wave plates were installed during OMEGA's upgrade to 351-nm, 60-beam, 40-TW capability in 1994. Notably, a few LCP devices installed at that time are still functional after 25 years of service. Such LC devices offer large-aperture scalability (200 mm), high optical quality and contrast with low insertion loss, precision retardance (tune by blending), broad angular tolerance, and excellent laser-damage resistance ($>30 \text{ J/cm}^2$, 1 ns, 1053 nm). In certain operational environments (e.g., such as in vacuum or where accidental fluid leakage can have additional adverse consequences), liquid materials are generally deemed unsuitable. Glassy liquid crystals (GLC's) offer the unique optical properties of LMM LC fluids, but the anisotropic ordering of LC phases is frozen into the solid state through vitrification. As a result, GLC's offer significant additional advantages such as resistance to mechanical disturbance, elimination of fluid leakage risk, and alleviation or reduction in the need for thick substrates.

To explore the potential benefits of GLC materials, this work is focused on designing high laser-induced–damage threshold (LIDT) GLC materials for large-aperture polarization control/beam-smoothing optics that could replace current LMM LC devices on OMEGA as well as offering the potential for use in other inertial confinement fusion (ICF)-class laser systems in future upgrades. A recent detailed study of LIDT in LC's performed at LLE indicated that reducing the absorption edge of LC materials significantly improves their damage thresholds for high-peak-power laser applications.¹ Based on that effort, a series of increasingly saturated (less aromatic) GLC's was synthesized with the goal of preserving their desirable optical and morphological properties while improving their endurance under irradiation by UV laser pulses and continuous-wave (cw), broadband UV light. This study explores an array of materials, ranging from an unsaturated aromatic GLC to highly saturated GLC's. The highly saturated GLC's show promising damage resistance under both irradiation conditions and offer considerable potential for application in both high-peak-power laser systems and aerospace optical systems.

The GLC material synthesis effort was complemented by examining the LIDT of these materials using both classical 1-on-1 and *N*-on-1 tests (yielding the 1-on-1 LIDT and the *N*-on-1 LIDT) as well as a 100-shot protocol. The latter was introduced in order to probe the long-term behavior and stability of the materials under continued exposure. Each site was shot at a given fluence at 0.1 Hz (once every 10 s) for either 100 shots *or* until damage was observed. The highest fluence at which a site survives 100 shots was defined as the 100-shot LIDT for the sample. A series of GLC's with progressively lower unsaturation and lower UV absorbance were synthesized, in which a stable cholesteric GLC phase can be generated by quench-cooling on substrates coated with rubbed alignment layers. Table I provides details on the materials synthesized and their characteristic properties. Purification by semi-automated flash chromatography was employed to remove trace contaminants and significantly improve LIDT's as compared to conventional purification methods (e.g., crystallization or precipitation from organic solvents).

The robustness of these materials to high-peak-power laser pulses as a function of unsaturation is evidenced by the LIDT data shown in Table I for three GLC materials tested at the three Nd:glass laser harmonic wavelengths and 1- to 1.5-ns pulse lengths. Test samples of 22- μm -thick GLC films, which are within the useful thickness regime for LC polarization control devices, were melt processed on fused-silica substrates and quenched into unaligned, nearly isotropic states to examine the intrinsic damage

Table I: Properties and LIDT values of GLC materials synthesized with differing levels of saturation in their respective molecular structures under exposure to laser pulses at 351, 523, and 1053 nm having durations of about 1 ns, 1.2 ns, and 1.5 ns, respectively.

GLC material	Aromatic rings	Absorption cutoff	1-on-1 (J/cm ²) 351/532/1053 nm	N-on-1 (J/cm ²) 351/532/1053 nm	100 shot (J/cm ²) 351/532/1053 nm
GLC-Bz3ChN2R	10	340 nm	0.61/-/-	0.43/-/-	-/-/-
GLC-Bz3CholC5	1	310 nm	4.9/16.1/38.9	-/-/-	1.0/4.3/22.3
GLC-CCH3CholC3	0	250 nm	5.9/-/16.4	3.9/-/9.7	0.88/-/5.2

resistance of each material independent of alignment conditions. For this set of materials (and particularly at 351 nm), the LIDT increases as they become more saturated and have deeper UV absorption edges [Fig. 1(a)]. For example, at 351 nm the most-unsaturated material, GLC-Bz3ChN2R, has only a meager 1-on-1 LIDT of 0.61 J/cm², which rises to 4.9 J/cm² for the more-saturated GLC-Bz3CholC5 and reaches 5.9 J/cm² for the highly saturated GLC-CCH3CholC3. These results follow the observed trend for LMM LC's, where LIDT values increase with increasing saturation and decreasing absorption edges.¹ As was seen in that study, reducing the π -electron delocalization and increasing the effective material band gaps appears to be quite beneficial in increasing the LIDT in GLC materials as well. While this principle holds true within experimental error at 351 nm, more-subtle details in the vibronic and electronic structures could potentially account for the different ordering seen in the 1053-nm LIDT values.

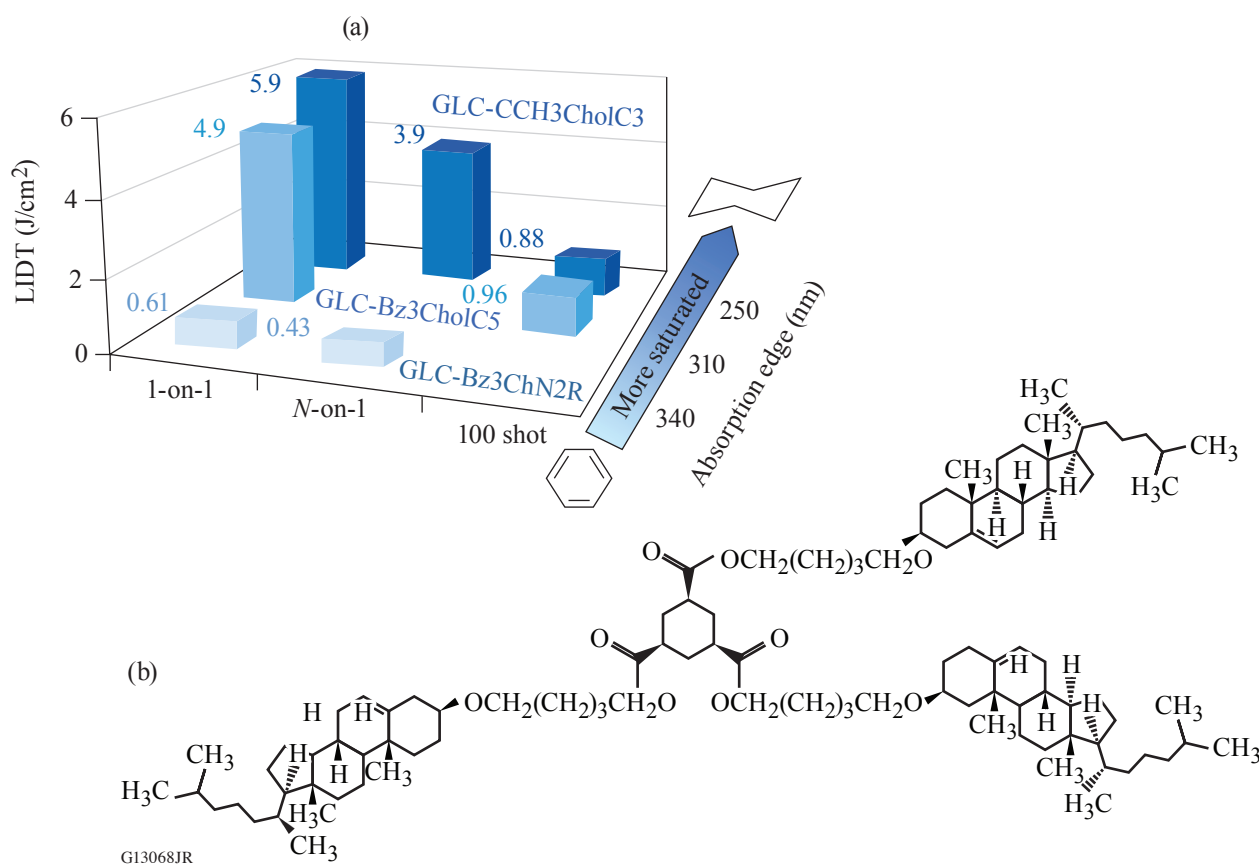


Figure 1

(a) LIDT values for 22- μ m-thick films of highly purified GLC materials sandwiched between two fused-silica substrates obtained with 351-nm, 1-ns pulses; (b) the molecular structure of GLC-Bz3CholC5.

Among this set of GLC materials, GLC-Bz3CholC5 shows the best overall performance for high-peak-power laser applications, particularly in the infrared (IR) spectral region, where its 100-shot LIDT was over 20 J/cm^2 . These results make GLC-Bz3CholC5 a promising candidate for use in polarization control or isolation elements for near-IR (NIR) lasers. A key behavior that remains of issue in all GLC materials synthesized to date is the degradation of the LIDT with continued exposure to laser pulses. This effect is observed for all damage-testing wavelengths, but it is more prominent for UV pulses. Specifically, the reduction of the 100-shot LIDT at 1053 nm in the best-performing GLC-Bz3CholC5 is approximately twofold compared to the 1-on-1 LIDT at the same wavelength, but it is about fivefold at 351 nm. To further understand the cumulative effects in the laser-damage behavior of these GLC's and explore them for use in cw UV environments, such as in aerospace applications as optical elements,² GLC films were subjected to broadband UV irradiation at 250 to 400 nm and their absorbance spectra were monitored as a function of exposure time. These exposure conditions increased the absorption coefficient in the UV region, which in turn contributes to lowering the LIDT values. Further understanding and development of mitigation strategies for this effect will be the subject of future work.

In conclusion, a series of increasingly saturated GLC's synthesized in multigram quantities and evaluated for their laser-induced-damage behavior at wavelengths relevant to OMEGA operations (1053 nm, 532 nm, and 351 nm) demonstrated promising performance. Reducing the π -electron delocalization in GLC molecules by employing saturated pendants, along with unconventional materials purification techniques, results in GLC's with improved LIDT's with 1-on-1 thresholds as high as 5.8 J/cm^2 at 351 nm, 16.1 J/cm^2 at 532 nm, and 38.9 J/cm^2 at 1053 nm. The results also demonstrate the potential for fabricating single-substrate GLC optical elements, especially for NIR laser applications where the 100-shot LIDT exceeds the current operational fluence of major ICF-class laser systems.

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1. T. Z. Kosc *et al.*, *Sci. Rep.* **9**, 16435 (2019).
2. E. Otón *et al.*, *IEEE Photonics J.* **7**, 6900909 (2015).