Growth of Large $CsLiB_6O_{10}$ Crystal from Li-Poor Self-Flux

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$CsLiB_6O_{10}$ (CLBO) is one of the key nonlinear optical crystals for generating deep-ultraviolet (DUV) light below the wavelength of 300 nm, and has already been put to practical use in coherent DUV illumination sources for semiconductor inspection. For next-generation advanced laser processing applications, output degradation occurred in the crystal is a critical issue associated with power scaling and long-term operation. In this research, we have investigated self-flux composition to produce high-quality CLBO crystals [1] and have grown large CLBO crystals with the weight of over 1 kg from Li-poor self-flux solution.

Three types of self-flux composition were examined: conventional B-poor flux, B-rich flux, and Li-poor flux. The starting materials were prepared from a mixture of Cs_2CO_3, Li_2CO_3, and H_3BO_3. After dissolving these materials in pure water, we charged the dried growth materials in a platinum crucible of 100 mm diameter. The crystal growth was conducted by Top Seeded Solution Growth (TSSG) method. The average growth rate along the *a*-axis was controlled to about 3 mm/day by adjusting the furnace temperature. The grown crystals with weight of about 100 g are free from cracks and macro inclusions. By using the accelerated UV-induced degradation test, the CLBO crystal grown from Li-poor flux exhibits relatively longer lifetimes than crystals grown from the other self-flux compositions.

Large CLBO crystals were grown from the Li-poor flux using a six-zone resistance heating furnace and a platinum crucible with a diameter of 200 mm and a height of 185 mm. A transparent CLBO crystal with dimensions of 161 mm (*a*) × 90 mm (*c*) × 65 mm (*a'*) and a weight of 1020 g was successfully grown for a period of 23 days. The average growth rate along the *a*-axis was about 3.6 mm/day. The weight is about three times heavier than that of conventional crystal. The large ingot enables to fabricate large-aperture devices for high-power DUV laser system. By using a device with an element size of 13 mm × 13 mm, 15-mm length (θ=61.9°, ϕ=45.0°), we have successfully demonstrated over 5,000-hour continuous operation of picosecond 266-nm DUV power of 10 W [2]. And by using a device with an element size of 16 mm × 16 mm, 15-mm length (θ=61.9°, ϕ=45.0°), we have also demonstrated over 10,000-hour continuous operation of picosecond 266-nm DUV power of 20 W [3]. After tuning the temperature distribution in the crucible, a transparent CLBO crystal with dimensions of 160 mm (*a*) × 103 mm (*c*) × 87mm (*a'*) and a weight of 1519.3 g was also grown for a period of 27 days. The average growth rate along the *a*-axis was about 2.8 mm/day. By using the large ingot, we fabricated the largest optical device of CLBO with dimensions of 70 mm × 70 mm × 15mm for second harmonic generation of high-energy infrared laser.

References

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