Yb:CaGdAlO4 (CALGO) for High Power Ultrafast Laser Applications

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Yb-doped $CaGdAlO_4$ (CALGO) is a very suitable material for high power ultrashort pulse laser applications. Its outstanding properties comprise a high thermal conductivity and an inhomogeneously broadened Yb-emission band caused by the statistical distribution of the Ca^{2+}- and Gd^{3+}/Yb^{3+}-ions on the same lattice site. Hence, it offers the ideal combination of glassy and crystalline material properties enabling it to overcome the previous limitations where one usually had to choose between high thermal conductivity (→ high power, crystal) and short pulses (→ statistical dopant distribution, glass).

Beginning in 2006, there have been repeated attempts to grow Yb:$CaGdAlO_4$ crystals in good optical quality for the said purposes. The state-of-the-art material was of a deep yellow to orange coloration while it was expected to be colorless and usually had clouds of small (2 µm to 5 µm) inclusions to it (Fig. 1).

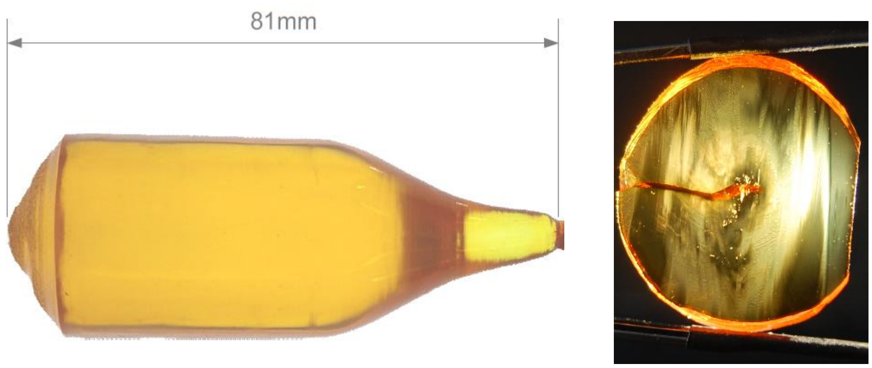
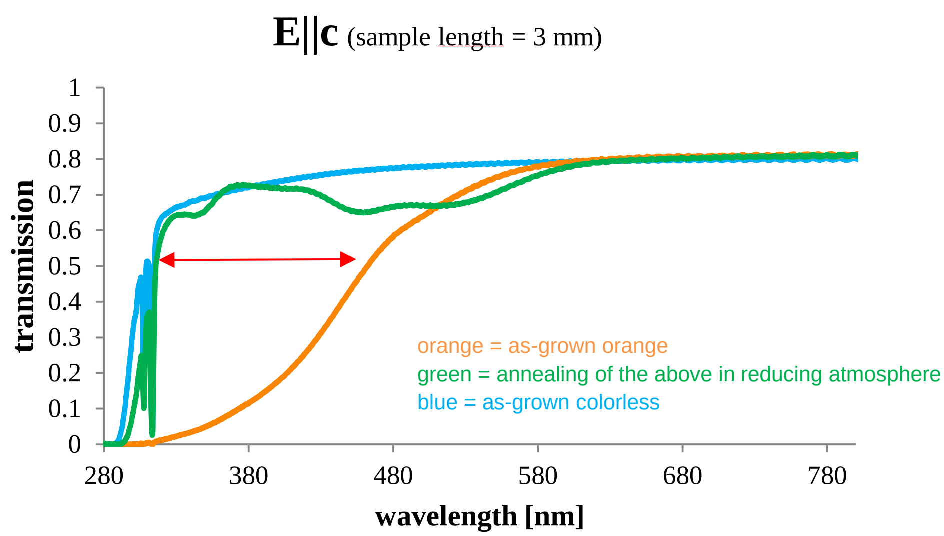


Fig. 1 As-grown Yb:$CaGdAlO_4$ crystal from 2006 (left). Typical appearance scheme of cloudlike inclusions of 2 µm to 5 µm size (right). Diameter of crystal sample 25 mm.

Since the inclusions scatter incident light and the color centers would cause additional unwanted absorption, both were expected to deteriorate laser efficiency and beam quality. Hence, the target was the elimination of these negative influence factors.

Up to 2013, various growth experiments were conducted only to find little impact on the crystal appearance when altering growth parameters such as pulling speed and rotation rate. It was learned, that an annealing treatment in a reducing atmosphere could remove the color in large part (Fig. 2, 3) while exposure to sunlight still led to a partial recoloration within days. This encouraged further studies to find a way to intrinsically remove the cause of coloration so that the as-grown material would not be prone to recoloration which would render a reducing treatment useless.

Fig. 2 Yb:$CaGdAlO_4$ boule after 12 h annealing treatment under reducing atmospheric conditions.



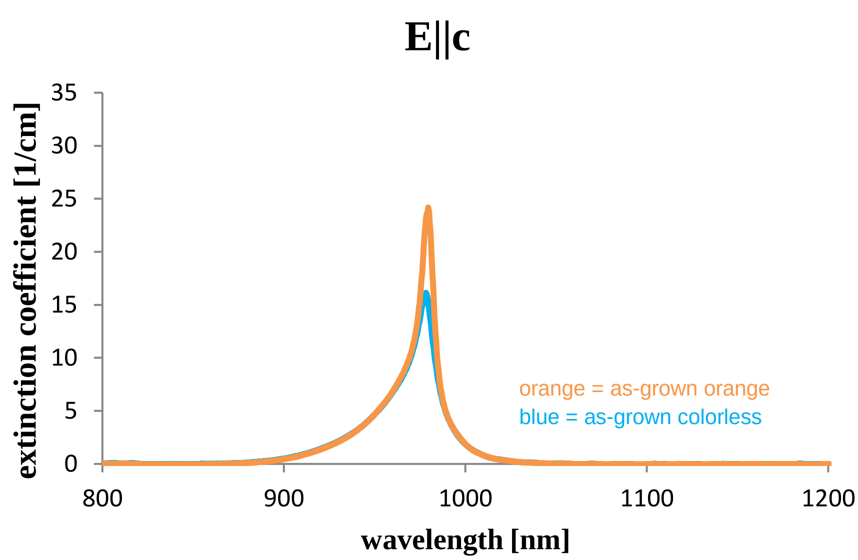


Fig. 3 Above: comparison of spectra from as-grown orange material, the same after an annealing treatment under reducing conditions and as-grown colorless material. Note the extended UV transparency range in the colorless material.
Below: extinction coefficients around the pumping wavelength. Note the increase in the colorless material.

Beginning in January of 2013, the above topic was subject to a PhD thesis[i] done at what was then FEE GmbH, now Electro-Optics Technology GmbH. From that point on to 2017, the major progresses on crystalline quality were made. Experiments included changes of the insulation setup (and thus thermal gradients), growth atmosphere and growth direction, but the most significant changes could be achieved by variations in the melt stoichiometry. The chemical analysis of the inclusions shows that this is mainly a matter of the molar ratio of Ca/Gd in the melt.

The coloration could be permanently removed and it was shown that there is a link between color depth and scattering center density (Fig. 3, 4). When exposed to daylight or moderate UV sources, no change in color could be observed any longer so that there should be an intrinsic mechanism which prevents the color centers from forming and that does not exist in the annealed samples. Experiments carried out by then High Q Laser GmbH in Rankweil, Austria, now part of Spectra-Physics[ii], could prove that the laser performance of colorless Yb:$CaGdAlO_4$ material is in fact 2 – 3 times better than its orange precursor (Fig. 5). Moreover, tests with different scattering center densities showed their influence on laser behavior to be comparably small.

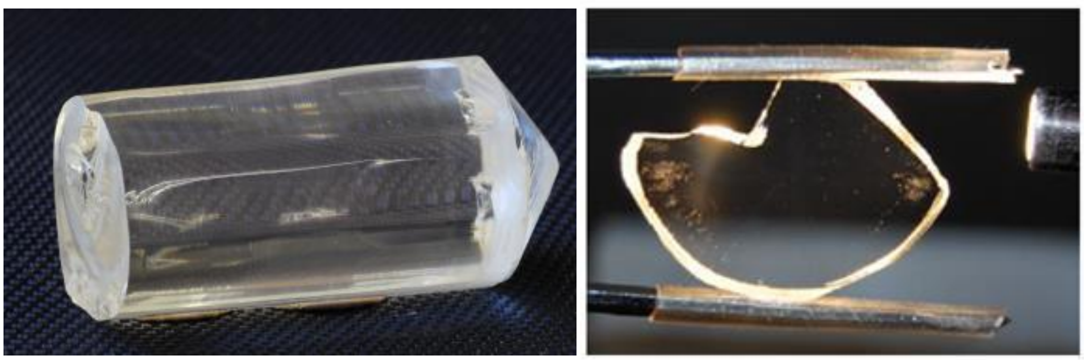
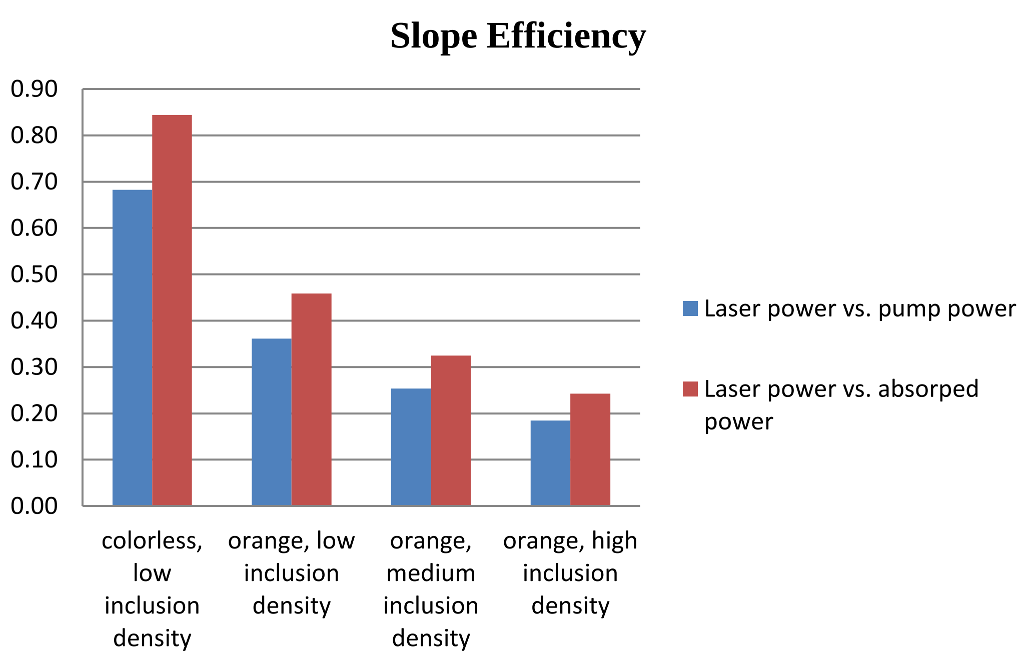


Fig. 4 Colorless as-grown Yb:$CaGdAlO_4$ crystal from 2017. Current state-of-the-art material.



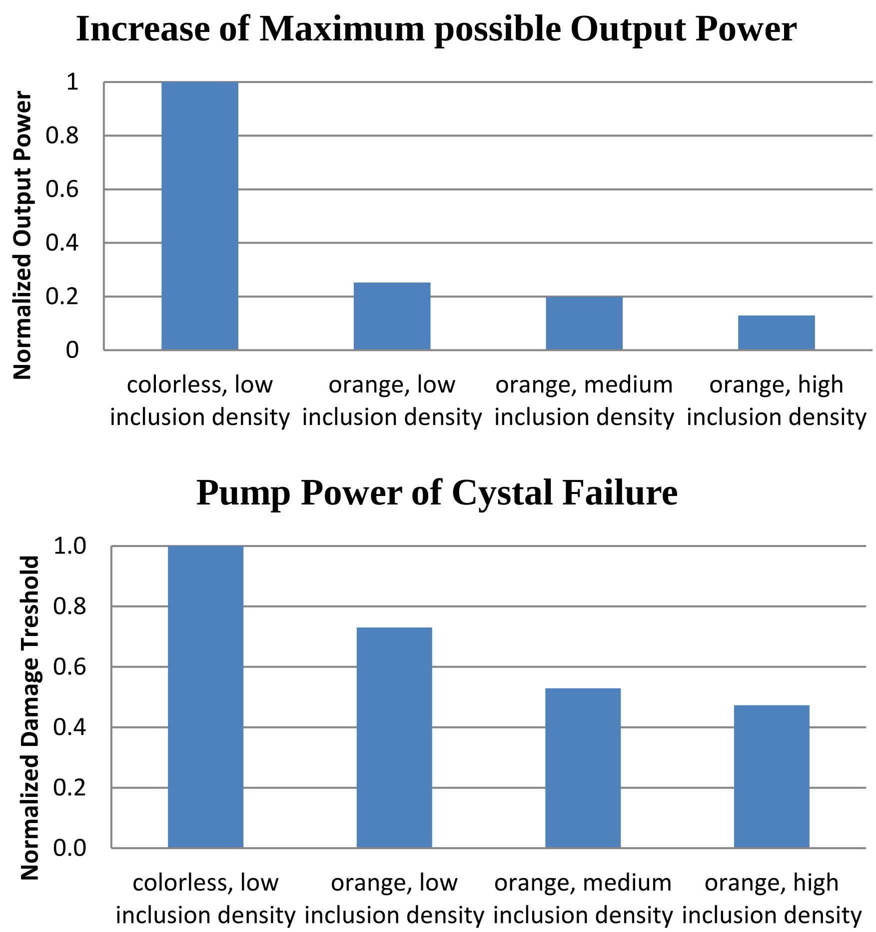
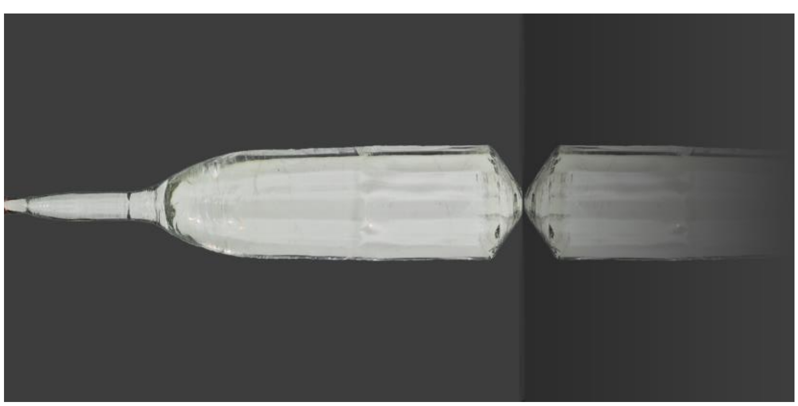


Fig. 5 Figures of merit for colorless Yb:CALGO material and orange material with different scattering center densities. Note the large influence of the crystal color while the effect of scattering is rather small on the laser performance.

Today, high quality Yb:CaGdAlO_4 is available off the shelf with its supreme performance in many different high power ultrafast laser systems always on display. There is still ongoing development and in 2019, the clearest material ever grown by Electro-Optics Technology GmbH is ready to show yet another increase in laser power and efficiency.



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__References:__

[i] **Liebald, C., “Yb-dotierte Ultrakurzpuls-Lasermaterialien mit K_2NiF_4-Struktur – Züchtung und Verbesserung der Kristallqualität“, PhD Thesis, University of Mainz, Germany (2017).**

[ii] **Aus der Au, J. and Kemnitzer, M., Testing Collaboration with FEE GmbH/EOT GmbH, Spectra-Physics, Rankweil, Austria (2013 – 2017).**