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Direct ink write fabrication of transparent ceramic gain media

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ABSTRACT

Solid-state laser gain media based on the garnet structure with two spatially distinct but optically contiguous regions have been fabricated. Transparent gain media comprised of a central core of $Y_{2.97}Nd_{0.03}Al_{5.00}O_{12.00}$ (Nd:YAG) and an undoped cladding region of $Y_3Al_5O_{12}$ (YAG) were fabricated by direct ink write and transparent ceramic processing. Direct ink write (DIW) was employed to form the green body, offering a general route to preparing functionally structured solid-state laser gain media. Fully-dense transparent optical ceramics in a "top hat" geometry with YAG/Nd:YAG have been fabricated by DIW methods with optical scatter at 1064 nm of <3%/cm.

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1. Introduction

Solid-state laser gain media has been dominated for over sixty years by single crystal growth methodology [1]. Over the last two decades, transparent ceramics have emerged as an alternative pathway for the fabrication of solid-state laser gain material with improved mechanical, thermal, and optical properties [2–5]. Transparent ceramics are being developed for various applications such as transparent armor, lighting fixtures, scintillators, IR windows and missile domes [6,7]. Advantages of transparent ceramics over single crystals include their rugged mechanical properties due to their polycrystalline microstructure, formability with higher doping concentrations and better doping uniformity, since they are fabricated via sintering rather than from the melt, as well as the ability to manufacture complex and large contiguous shapes. Secondarily, these materials can be more cost effective in comparison to their single crystal counterparts, as they do not require slow solidification from the melt in order to provide large transparent boules. Transparent ceramics possess various advantages over conventional optically transparent materials such as glasses, in that they are crystalline – important for obtaining narrow spectroscopic linewidths with rare-earth dopants, such as Nd^{3+}. While single crystal and glass laser gain media are advanced and well-developed technologies, transparent ceramics offer functional opportunities not readily attainable with structures grown from the melt.

Previous efforts to produce structured ceramic laser gain media have relied on assembly of multiple layers at the green body stage [8–10]. In this contribution, we describe the fabrication of a Nd^{3+} dopant profile that is concentrated in the center of a Nd:YAG laser rod as a "top-hat" step profile, (or in the future as a smooth, graded profile), to increase the pump efficiency by a factor of 2× while stabilizing the desired laser mode profile in the transverse electromagnetic (TEM_{00}) mode. Ceramics processing enables the fabrication of a spatially-controlled chemical composition at the stage of green body fabrication, formed via direct ink write (DIW). We are using 3D DIW printing techniques [11–14] to fabricate compositionally-controlled green bodies, subsequently sintered to transparency for use as laser gain media.

As shown in Fig. 1a, a laser rod with a "top hat" Nd^{3+}-doped gain region, in principle, has built-in laser mode-control within the structure that better matches the TEM_{00} laser mode than a uniformly-doped Nd:YAG rod, while the TEM_{01} and other higher-order modes are not favored, since no Nd^{3+} dopant is present beyond 0.1 cm in the cladding region in this design. (The details of this standard laser calculation are summarized in the Appendix.) Fig. 1b shows that the predicted output power for the TEM_{00} mode in a Nd:YAG ceramic with a "top hat" dopant profile is favored, compared to the first higher order mode (TEM_{10}) shown for comparison, which has a higher threshold and a reduced slope efficiency. We note in passing that for a typical *highly multimode* laser, doped at 0.9 at.% and having a loss level of 0.3%/cm (which are typical specifications for commercial crystalline laser rods), the

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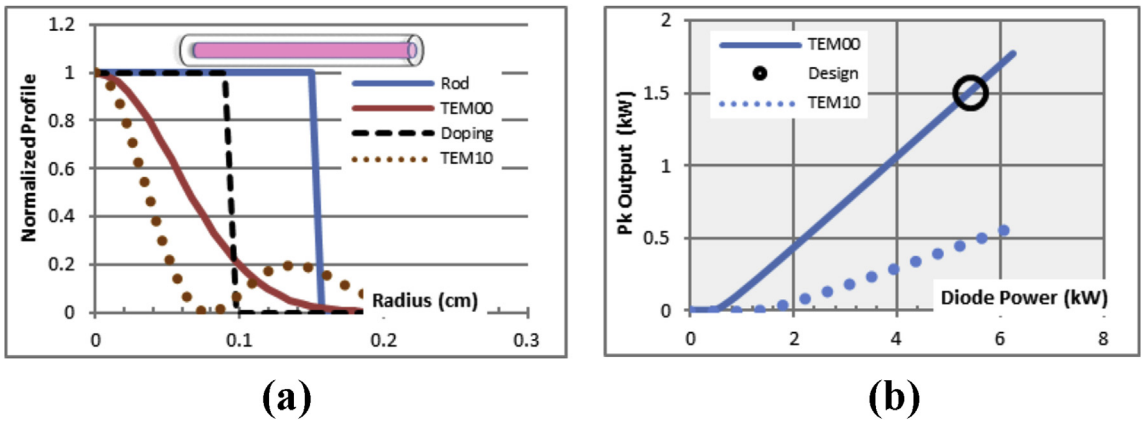

Fig. 1. **(a)** Normalized profile of the rod radius (blue line), "top hat' doping profile (black dashed line), TEM_{00} intensity profile (red line), and TEM_{10} profile (red dots). **(b)** Peak output power as a function of pump diode power corresponding to the TEM_{00} laser mode (solid blue line), and TEM_{10} laser mode (blue dots). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

peak power at the design point is 2.27 kW at the circled design point on the Figure – which is only modestly higher than the modeled single-mode laser. A final issue to be addressed in the design of a practical "top hat" ceramic laser rod with a Nd-doped core or gradient involves the change in refractive index induced by the Nd doping of $\Delta n = 0.00043$/at.%, as reported by Zelmon et al. [15] and reproduced by us. At 2% Nd-doping, this would amount to 50 waves of distortion between the core and the clad for a 6 cm rod, which however can be compensated for by way of the addition of ~10% Lu-dopant to the "undoped" YAG region, (controlled to ±0.1% doping accuracy).

Previous efforts to build optical structures with compositional gradients have involved assembling pressed and layered green bodies (Konoshima, Japan [16]) and using diffusion bonding to join carefully cut and polished single crystal optical components (Onyx Optics, Pleasanton, USA [17]). At LLNL, "top hat" laser rods with Nd^{3+}-doped core and undoped cladding have been fabricated via composite green body assembly (Fig. 2). However, radially-graded green bodies are difficult to prepare by this approach, as smooth gradients are not easily achieved. In this paper we report on our efforts to adapt DIW methods to the requirements for a functionally graded transparent ceramic.

2. Experimental procedure

2.1. Powder characterization

Fabrication of high quality, fully-dense and transparent optical ceramics requires high purity precursor powder. Nanoparticles with the compositions $Y_3Al_5O_{12}$ and $Y_{2.94}Nd_{0.06}Al_5O_{12}$ (YAG/YAG:Nd) were fabricated via flame spray pyrolysis (FSP) by Nanocerox and then calcined in air between 900 and 1200 °C. The morphology of the calcined YAG nanopowders were analyzed via bright-field transmission electron microscopy (TEM) using a FEI Titan 80-300 S/TEM with an operational voltage of 300 kV. X-ray diffraction (XRD) performed using the XRD-Bruker AXS D8 Advance, a Cu anode diffractometer ($K\alpha_1 = 0.1540598$ nm). The X-ray diffraction patterns were displayed via Bruker AXS EVA data acquisition software. The samples were prepared by dispersing the nanopowders onto silicon single crystal wafers with acetone using a step size of 0.059482 from 16° to 60° diffraction angle range. The surface area measurements of the YAG and Nd:YAG nanoparticles were determined by Brunauer, Emmett, and Teller (BET) analysis using a Micromeritics ASAP 2020 surface area and porosity analyzer.

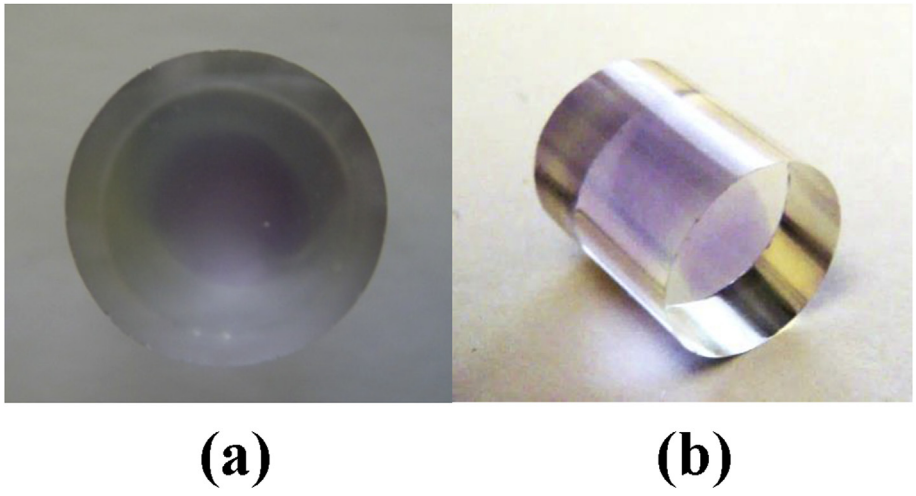

Fig. 2. "Top-hat" transparent ceramic with Nd^{3+} doped core and undoped cladding formed by green body assembly methods at LLNL **(a)** top-view and **(b)** side-view.