Design and characterization of Yb and Nd doped transparent ceramics for high power laser applications: recent advancements

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ABSTRACT

We report a review on our recent developments in Yttebium and Neodymium doped laser ceramics, along two main research lines. The first is the design and development of Yb:YAG ceramics with non uniform doping distribution, for the management of thermo-mechanical stresses and for the mitigation of ASE: layered structures have been produced by solid state reactive sintering, using different forming processes (spray drying and cold press of the homogenized powders, tape cast of the slurry); samples have been characterized and compared to FEM analysis. The second is the investigation of Lutetium based ceramics (such as mixed garnets LuYAG and Lu_2O_3); this interest is mainly motivated by the favorable thermal properties of these hosts under high doping. We recently obtained for the first time high efficiency laser emission from Yb doped LuYAG ceramics. The investigation on sesquioxides has been focused on Nd-doped Lu_2O_3 ceramics, fabricated with the Spark Plasma Sintering method (SPS). We recently achieved the first laser emission above 1 W from Nd doped Lu_2O_3 ceramics fabricated by SPS.

Keywords: Laser ceramics, Nd:YAG, Yb:YAG, Lu_2O_3 sesquioxide, Layered ceramics, SPS method.

1. INTRODUCTION

Important advances have been recently achieved in the field of solid state laser materials through the development of transparent ceramics. The first breakthrough was obtained by A. Ikesue[1], who obtained substantially equivalent laser output from Nd^{3+}:YAG crystal transparent ceramic, overcoming the problems related to scattering. Laser performances of commercial YAG ceramics are currently fully comparable with their crystalline equivalents[2-5]. In our recent work on in Yb and Nd doped laser ceramics we aimed our investigations along two main lines. The first is the design and development of Yb:YAG ceramics with non uniform doping distribution, for the management of thermo-mechanical stresses and for the mitigation of ASE and re-absorption processes. Layered structures have been produced by Solid State Reactive Sintering (SSRS), using forming processes such as spray drying and cold press of the homogenized powders, or tape cast of the slurry. Composite structures have been characterized and compared to FEM analysis[6]. The second line is the investigation of ceramics with new compositions based on Lutetium, with Yb and Nd doping. The interest on Lu-based materials as laser hosts is mainly motivated by their thermal properties: in hosts such as LuAG, LuYAG and Lu_2O_3 doped with Nd^{3+}, Er^{3+}, Yb^{3+} the thermal conductivity remains fairly constant[7] even at high doping level, because of the small atomic weight difference between Lu^{3+} and the substituting ion. Conversely, the thermal conductivity of YAG significantly decreases with doping, due to the larger difference between the atomic weight of the doping ion and of the Y^{3+} . These features are very important in high power laser scaling up. A higher thermal conductivity favors the transport of heat from the active medium pumped regions to the heat sink and reduces peak temperatures and temperature

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gradients, reducing thermally induced stresses and allowing higher pump power densities on the active medium. All these advantages can improve the performances of High Power/High Energy Diode Pumped ceramic sources[8,9]. In this framework we also investigated mixed garnets ($(Lu_xY_{1-x})_3Al_5O_{12}$, LuYAG) doped with Yb, obtaining for the first time laser emission from these ceramics, with a high efficiency and relatively broad tuning range[10,11]. The investigation on Nd-doped Lu_2O_3 ceramics, fabricated at the Tohoku University (Japan) by the Spark Plasma Sintering (SPS) method[12-14] resulted in the first extraction of powers above 1 W.

2. LASER CERAMICS WITH NON-UNIFORM DOPING DISTRIBUTIONS

The control in the distribution of the doping enables to mitigate the thermal and thermo-mechanical effects (e.g. thermal lens, stress-induced depolarization, surface deformations) deriving from the laser pumping process. These effects can degrade the performance of the laser source, eventually leading to a catastrophic failure of the laser active element under high pump power densities. Finally, a suitable non uniform doping distribution can reduce Amplified Spontaneous Emission (ASE) effects with respect to an uniform doping. To evaluate this approach we analyzed both numerically and experimentally end-pumped active media a layered, non uniform doping distribution.

2.1 Design and numerical analysis of Yb:YAG end-pumped layered systems

To study the behavior of ceramic layered structure with different doping levels we developed a 3D numerical analysis of the slab optical properties as a consequence of the thermal load induced by the pump process, based on on Finite Element Mesh (FEM) analysis[6]. The model allows both longitudinal and transverse variations of the parameters, enabling to estimate thermal distributions and thermo-mechanical stresses in spatially structured active media.

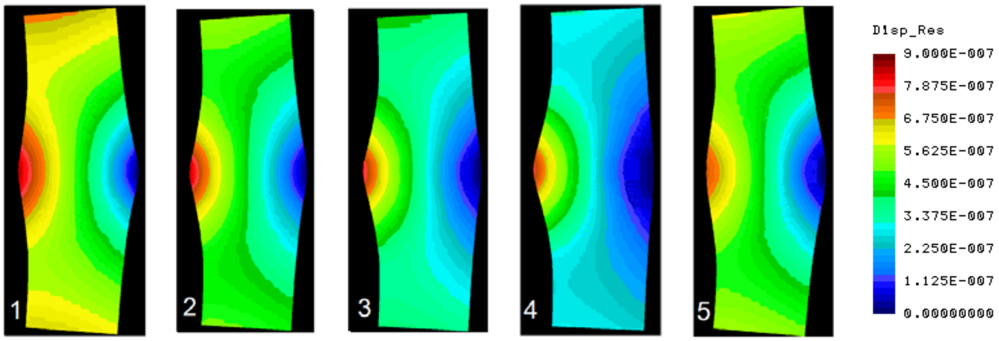

Figure 1. Surface deformation in an axial cross-section for five differently structured layered samples (see Ref.6) under the same thermal load in a ring cooling geometry.

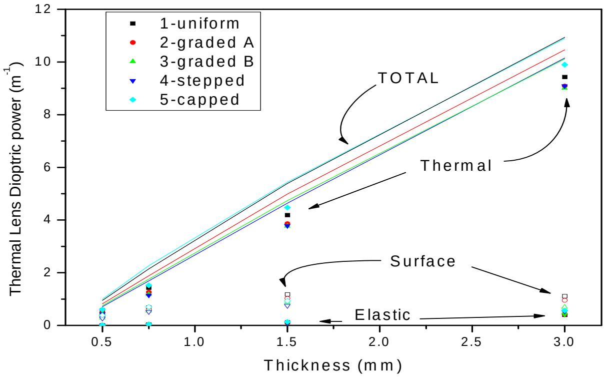

Figure 2. Thermal lens contributions (thermal, stress induced and surface deformation) as a function of the thickness for a given diameter of the active volume.

We concentrated on end pumped disc-like systems with longitudinally varying doping concentrations, both with ring-cooling and face-cooling. Our analysis included uniform active media, graded active media, stepped active media (with doped-undoped interfaces). The FEM thermo-mechanical simulation produces the internal temperature and stress distribution. The effects on the refractive index (thermal lens and depolarization estimations) are determined with a tensor post-processing calculation. Fig.1 shows an example of the deformation of five differently doped samples under the same total thermal load. Interestingly, a longitudinal doping variation doesn't affect the thermal lens as long as the cooling is substantially radial[6] (differently from disk or slab systems with high aspect ratios) as summarized in Fig.2.

2.2 Experimental characterization of Yb:YAG end-pumped layered systems

The numerical simulations were validated by means of experimental measurements of thermal lens (TL) on uniform and structured samples produced with the spray-drying method. All the experiment refer to a single sided standard end-pumping scheme[15]. The layer thicknesses and doping levels of the tested samples are summarized in Table 1. The TL effect in the different samples was characterized using the setup shown in Figure 3. This was done both on lasing and non lasing samples, to show possible differences in the thermal load. The samples are end pumped by a fiber coupled diode laser (pump spot 480 µm diameter @1/e^2). The TL is evaluated by measuring the wavefront distortion of a probe beam emitted by a HeNe laser, using a Shack-Hartmann sensor. The dioptric power of the TL is proportional to the absorbed power, with a linear coefficient *C* reported in Table 5. A more detailed description of the TL evaluation and its dependence on the probe beam aperture is given elsewhere[15]. TL measurements show little differences between the various samples, given the substantially radial heat flow of our cooling geometry. These experimental tests substantially confirm the predictions of our numerical analysis. This validation allows us to our model it to other geometries and doping distributions. This will be illustrated in the following section for a second case study.

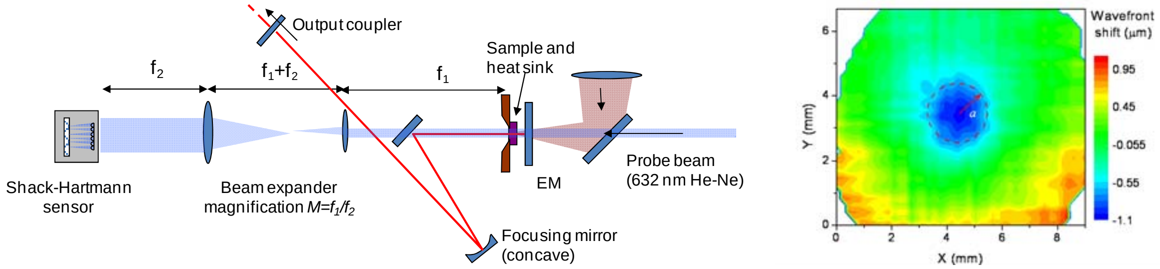

Figure 3. (Left) Experimental set up for the thermal lens measurements; (Right) Example of wavefront deformation map.

Table 1. Thermal lens dioptric power Coefficient C (dioptric power per Watt of absorbed pump radiation).

Sample	Doping levels	Layer Thickness [mm]	***C (Laser on)***	***C (Laser off)***
Uniform (Ceramic)	10 %	1.25	0.889	0.919
Uniform (Crystal)	8%	2	0.959	0.988
Stepped (Ceramic)	(0,10,0)%	(0.6-1.2-0.6)	0.870	1.003
Capped (ceramic)	(0,10)%	(1.6-2.0)	1.138	1.166

Table 2. Parameters of the samples used in the efficiency comparison experiments. SD. Samples made by cold pressing of spray dried powders; TC: samples made by tape casting and thermal compression.

Sample n.	Yb doping %*at.*	Total thickness (mm)	Individual layers thickness (mm)	Laser slope efficiency
SD-1	0-10	3.4	1.5 (0% Yb) -1.9 (10 % Yb)	58.1 %
SD-2	1-3-5-7	2.0	0.5	19.5 %
TC-1	0-10	1.9	1.0 (0% Yb) - 0.9 (10% Yb)	52.2 %
TC-2	1-3-5-7	2.0	0.5	43.8 %

Laser efficiency measurements have been performed to compare samples produced with different methods. Table 2 summarizes the properties and the laser efficiencies of samples produced at ISTEC with the Spray-Drying (SD)[16] and Tape-Casting (TC)[17] methods. These results show a substantial equivalence of the two methodologies in the case of a small number of layers (2) while they indicate a relevant quality degradation when a larger number of layers is used. Tape casting seems more suited for this kind (large number of thin layers) of devices.

2.3 Numerical analysis of Yb:Lu_2O_3 layered systems

We repeated a numerical analysis campaign for samples with the same geometries but based on different hosts. YAG was compared to Lu_2O_3 and Sc_2O_3 which, as mentioned above, present higher thermal conductivity than YAG, under relatively high Yb^{3+} doping levels (see table 3). Therefore for Lu_2O_3 hosts we obtained lower maximum temperatures and lower deformations (thus thermal lens and depolarization), as summarized in Fig.4.

Table 3. Parameters of the different hosts compared in our numerical simulations.

Parameter	YAG	Lu_2O_3	Sc_2O_3
Doping	10%, 0%	4.5%, 0%,	3.0%, 0%
Absorption coeff. (cm^{-1})	5 (10% Yb),0 (0% Yb)	5 (4.5% Yb), 0 (0% Yb)	5 (3.0% Yb), 0 (0% Yb)
Thermal Cond. [W/(mK)]	6.7 (10% Yb), 10.7 (0% Yb))	11.9 (4.5% Yb), 12.5 (0% Yb)	7.0 (3.0% Yb), 16.5 (0% Yb)
Thermal exp. Coeff. [K^{-1}]	8×10^{-6}	5.5×10^{-6}	6.7×10^{-6}
Young Modulus	0.280×10^{12} Pa	0.178×10^{12} Pa	0.221×10^{12} Pa
Poisson Number	0.28	0.28	0.28

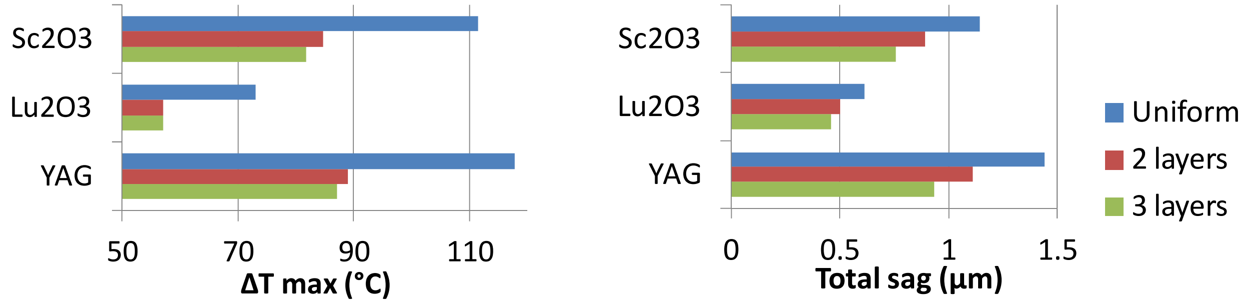

Figure 4. Summary of max internal temperature increase and max sag of layered samples based on different hosts.

3. OTHER HOST FORMULATIONS AND PREPARATION PROCEDURES

Our recent activities in the developments of laser ceramics also included the investigation on alternative material preparation routes, and different formulations. In particular, were investigated the use of laser ablated nanopowders as a starting materials for the production of Nd:YAG ceramics. The other formulations under study included Nd:Lu_2O_3 fabricated by means of the SPS method, and mixed garnet with formulation Yb:LuYAG.

3.1 Nd:YAG ceramics produced from laser ablated powders

Transparent Nd:YAG ceramics were prepared at the Institute of the Electrophysics (Russia) by the SSRS method using nanopowders of 1 at.% Nd:Y_2O_3 and Al_2O_3 synthesized by laser ablation. A pre-calcining step and addition of tetraethyl orthosilicate were found crucial for the quality of the samples fabricated from such nanoparticles[18]. The transmittance of a 2-mm-thick Nd:YAG ceramic was 83.6% at 1064 nm, very close to the theoretical. Output power of 4.9 W (slope efficiency 52.7%) was obtained from Nd:YAG ceramic with quasi- CW end pumping at 805 nm (Fig. 5).

3.2 Nd:Lu_2O_3 ceramics produced by SPS

SPS is a new sintering technique that has been successfully applied to the fabrication of Lu_2O_3 transparent ceramics for laser applications. 1% Nd doped samples were prepared at the at the Tohoku University, Sendai (Japan) [19]. SPS is comparatively simple and it requires less time than the conventional sintering methods (usually tens of minutes instead of several hours) and lower processing temperatures[12, 13]. This is particularly attractive for sesquioxides because these materials are very difficult to grow as single crystal, due to their very high melting temperature (e.g. 2490 °C for Lu_2O_3). For these samples the sintering phase required about 90 min (plus cooling), with a peak processing temperature of 1450°. Laser output from these ceramics was already reported by us[20] using an experimental set up similar to that shown of Fig. 5 (a); recently, we obtained a maximum output power above 1 W with a maximum slope efficiency of 12.9% (fig. 5 c).

3.3 Yb:LuYAG ceramics

Besides the problem of the high processing temperature mentioned above, the fabrication of Lu-based hosts such as Lu_2O_3 and LuAG has the disadvantage of the high cost of the high purity Lu_2O_3 powder required for both the ceramics and the crystals; to overcome these problems, the mixed garnet resulting from solid solutions of LuAG and YAG, i.e. $(Lu_xY_{1-x})_3Al_5O_{12}$ (Lutetium-Yttrium Aluminum Garnet, LuYAG) was recently proposed [21], as crystals and ceramics. Due to the partial substitution of Lu with Y, it requires a lower quantity of Lu^{3+}, it has a lower melting point than LuAG and Lu_2O_3, and a fairly high thermal conductivity (7.8 W/m•K [22]). Recently, we have demonstrated for the first time the laser action for Yb:LuYAG ceramics with 15% Yb doping and various Y/Lu balances [23, 24]. Samples were prepared at the Shanghai Institute of Ceramics (SIC-CAS, China) by reactive sintering; a detailed spectroscopic analysis was carried out at the Institute of Physics of the Czech Academy of Sciences (Prague). Laser tests were carried out at INO-CNR using a longitudinally pumped cavity (Fig. 6). Remarkable laser emission results were achieved, with maximum output power of 8.2 W, 7.3 W and 8.7 W for Y/Lu balance 25/75, 50/50 and 75/25 respectively, at 1030 nm, with a maximum absorbed pump power of about 16 W; the slope efficiency and the optical-to-optical efficiencies approached or exceeded 60% and 50% respectively.

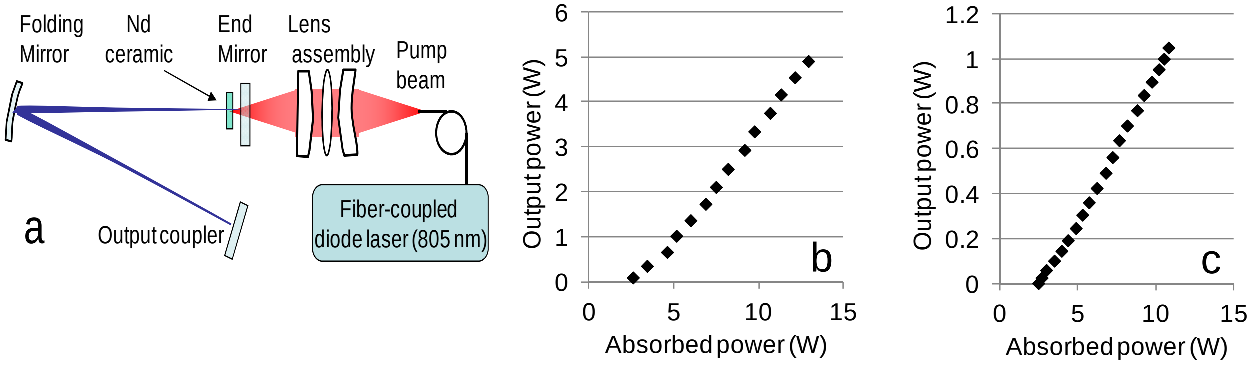

Figure 5. (a) Experimental set-up for the laser test on the Nd:doped ceramics. (b, c) Output power vs. absorbed pump power for the Nd:YAGand Nd:Lu$_2$O$_3$ (output coupler reflectivity respectively 80% and 98%). QCW pumping, duty factor 12.5%.

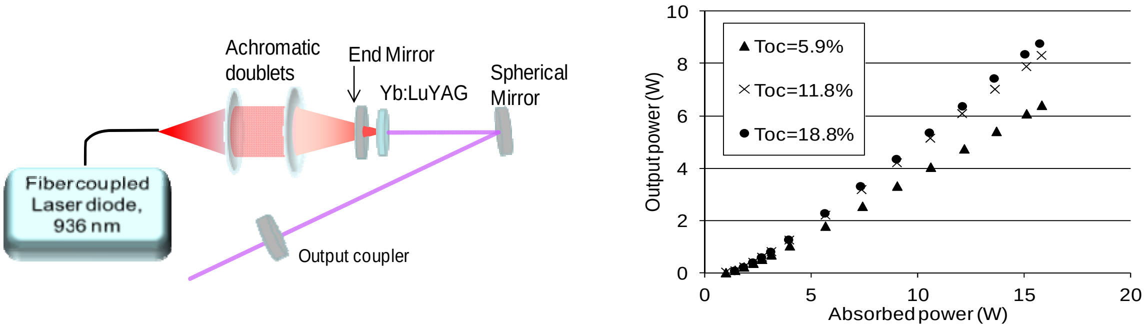

Figure 6. (Left) Experimental set-up for the laser test on the Yb:LuYAG ceramics, (Right) Output power vs. absorbed pump power for the sample with Y/Lu=75/25 (QCW pumping, pump duty factor 20%; Toc: output coupler transmission).

4. CONCLUSIONS

We have investigated, both numerically and experimentally, the performance of ceramic active media with various host compositions and with structured design. In particular end-pumped discs with a layered structure with varying doping level have been thoroughly analyzed. These structured ceramics appear promising in appropriate geometrical configurations, and thus are intended to be applied in the construction of High Power Diode Pumped Solid State Laser (DPSSL) or High Energy pulsed systems working in high repetition-rate pulsed regimes.

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