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<td><a href="http://hdl.handle.net/10220/10933">http://hdl.handle.net/10220/10933</a></td>
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Yb:LuAG laser ceramics: a promising high power laser gain medium

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Abstract: Yb:LuAG laser ceramics with different Yb3+ doping concentrations were successfully fabricated by using a solid-state reactive sintering method. SEM graphs demonstrate that the samples have a dense and pore-free microstructure. Based on the spectroscopic studies the ceramics have a large emission cross-section of $2.7 \times 10^{-20}$ cm$^2$ at 1030 nm emission peak. CW laser operation of the samples has given 7.2 W output power with 65% slope efficiency.

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OCIS codes: (140.3580) Lasers, solid-state; (160.3380) Laser materials.

References and links


Received 23 Jul 2012; revised 5 Sep 2012; accepted 9 Sep 2012; published 20 Sep 2012

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1. Introduction
Since the invention of the first laser by Maiman 50 years ago [1] there has been enormous progress on the laser science and technology. Nowadays lasers as an enabling tool have found widespread applications in almost all fields of the modern science and technology. On the other hand, as performance of a laser is essentially determined by the property of its gain medium, the search for better laser gain media to replace the existing ones has never stopped. Recently, the research on using the transparent ceramics as the laser gain host has attracted great attention. With the modern ceramics fabrication technology a polycrystalline ceramic could be made transparent with high optical quality. Compared with the single crystals, a polycrystalline ceramic laser material would have a number of advantages such as short production period, low production cost, and flexible composite structures [2]. Since Ikesue et al. first demonstrated efficient operation of a Nd:YAG ceramic laser in 1995 [3], the research and development on laser ceramics and ceramic lasers has been keeping in a fast pace. So far, a number of rare-earth ions doped ceramic laser gain materials have been successfully developed [4–8].

The rare-earth ytterbium (Yb) ions have a simple two-level energy structure and no upconversion, no excited state absorption, and no concentration quenching [9]. Therefore, the Yb-doped crystals as laser gain media have been previously extensively investigated. It has been shown that they have the remarkable features of high quantum efficiency (small quantum defect), very broad emission spectrum, and long fluorescent lifetime, which favor high power and ultra-fast laser applications. Recently, the Yb-doped YAG and sesquioxide ceramics have also been developed. High efficient and kilowatts operation of the Yb-doped lasers was reported previously [10–13].

In the high power solid-state laser operation there is unavoidably a large amount of heat generation. If it could not be efficiently removed, it could cause severe thermal problems, even lead to permanent laser gain medium damage. Therefore, selection on the laser gain host material becomes especially important for the high power lasers [14]. Traditionally, the Yb:YAG is widely used for the high power laser applications because it has a large thermal conductivity. However, it was found that as the Yb doping concentration in the YAG is increased, the thermal conductivity of the Yb:YAG drops significantly [11,15,16]. It affects severely the high power performance of the lasers.

To solve the problem, the solid solution of (Lu,Y)3Al5O12 (Lutetium Yttrium Aluminium Garnet) was proposed as an alternate Yb host. Kuwano et al. has previously reported the growth of solid solution LuYAG single crystal [17]. On the other hand, the end compound Lu3Al5O12 has high density (6.72 g/cm3), high thermal conductivity (9.6 w/m·K) [17], and a cubic structure that is optically isotropic. The atomic mass and the ion radius of Lu ion...
(174.97, 100.1 pm) are close to those of the Yb ion (173.04, 100.8 pm) rather than the Yttrium ion (88.9059, 104 pm). So in the high Yb doping concentration, the small mass difference between the host-cation and the dopant will cause less phonon scattering, which is the main reason for the dropping in thermal conductivity [15]. Although the thermal conductivity of the LuAG is slightly smaller than that of the YAG, the thermal conductivity of the Yb:LuAG becomes well above Yb:YAG when Yb doping concentration increases larger than 5% [15]. This property of Yb:LuAG attracted many researchers’ attention. Sumida et al. reported the first room temperature diode pumped Yb:LuAG single crystal laser in 1995 [18]. A. Brenier et al. reported the growth, spectroscopic, and laser properties of the Yb:LuAG crystal in 2006 [19]. K. Beil et al. reported a 5 kW Yb:LuAG thin disk laser with optical-to-optical efficiency of over 60% [11]. The first Q-switched Yb:LuAG microchip laser was reported in 2007 [20]. He et al. achieved mode-locking of a Yb:LuAG single crystal laser with ~7.6 ps pulse width [21].

However, there’s only a few reports on the Yb:LuAG ceramic lasers. Besides Xu et al. reported the operation of a Yb:LuAG ceramic laser for the first time [22], and very recently Nakao et al. reported a CW and mode-locked operation of a Yb:LuAG ceramics [23], no other researches on the fabrication of Yb:LuAG ceramics were reported, as the fabrication of high optical quality Yb:LuAG ceramics still remains a challenge.

Here we report on the fabrication and spectroscopic properties of high quality transparent Yb:LuAG polycrystalline laser ceramics. LuAG ceramics with various Yb-doping concentrations from 5% to 20% were successfully fabricated by using a solid-state reactive sintering method, and high power high efficient lasing operation of the fabricated Yb:LuAG ceramics was experimentally demonstrated.

2. Experimental procedures

The raw materials were high purity (>99.99%) Al₂O₃, Lu₂O₃ and Yb₂O₃ powders. The primary particle size of the Al₂O₃ powder was around 250 nm and the specific surface area of the powder was ~11.0 g/m². The primary particle size of the Lu₂O₃ and Yb₂O₃ powders were 60–80 nm and the specific surface area of the powders were ~10.2 g/m² for Lu₂O₃ and ~9.4 g/m² for Yb₂O₃. The detailed fabrication process of the Yb:LuAG ceramics is similar to that used for fabricating the Yb:YAG ceramics [24]. Briefly, the raw powders were weighed carefully according to the Yb:LuAG chemical stoichiometric composition. The different Yb doping concentrations were selected to be 5.0 at.% 10.0 at.% 15.0 at.% and 20.0 at.%, respectively. The powders were mixed with 99.99% analytical pure ethanol for ball milling. The mixed slurry was then ball milled using a planetary milling machine for about 15 h. SiO₂ as sintering aids were introduced by adding 0.5 wt.% tetraethyl orthosilicate (TEOS, Sigma-Aldrich, 99.999%). The milled mixtures were then dried in oven and sieved through a 100-mesh screen. After calcining at 800 °C for 3 h to remove organic component, the powders were then dry pressed in a stainless steel die at:15 MPa to form a pellets shape. The green body pellets were further cold isostatically pressed (CIPed) at 200 MPa. The relative green body density reached around:53% after CIP. The green body was finally sintered at 1790 °C for 12 h in a high temperature vacuum sintering furnace under a vacuum level better than 10⁻⁴ Pa to achieve the theoretical density. The sintered pallets were then annealed in air (1450 °C for 15 h) to remove internal stress and eliminate the oxygen vacancies. The transmission spectra of samples were obtained by a UV-VIS-NIR spectrophotometer (UV-3600, SHIMADZU) from 850 to 1100nm with a scan rate of 0.3 m/s. The room temperature emission spectra and the decay time of the samples were measured by a spectrofluorometer (Fluorolog-3, Jobin Yvon, Edison, USA) when the samples were excited by a 940 nm laser diode. For the fluorescent lifetime measurement, the fine grinded powder of the sample was dispersed in the glycerin solution to reduce the re-absorption effect on a certain level. The SEM graph was taken by a JEOL JSM-6360A SEM. To this end the samples were surface polished and thermally etched at 1500 °C for 3 hours to reveal the grain boundary.
3. Results and discussion

Figure 1 shows the microstructure of the Yb:LuAG polycrystalline ceramic samples. The SEM graphs for samples with Yb doping concentration of 5% (a), 10% (b), 15% (c) and 20% (d) are displayed. They all show an almost pore-free microstructure. The grain size decreases slightly with the increase of the Yb doping concentration. The samples have an average grain size of ~10 μm.

![Figure 1](image1)

Fig. 1. Microstructure of the polished and thermally etched Yb:LuAG polycrystalline ceramics with 5% (a), 10% (b), 15% (c) and 20% (d) Yb doping concentrations.

Figure 2 displays the room-temperature absorption spectra and a photo of the as fabricated Yb:LuAG ceramic samples. The samples are round pellets with 17 mm in diameter and 3 mm in thickness. The samples are mirror polished on both surfaces and appear blue in color before annealing. The color becomes darker as the Yb doping concentration is increased, which is due to the Yb$^{3+}$ formed during the vacuum sintering. After being annealed in air they become colorless and fully transparent. The absorption spectrum of the Yb:LuAG ceramics is similar in shape with that of the Yb:YAG ceramics. The highest absorption peak is at 968.4 nm. For the samples with 5%, 10% and 15% Yb doping concentration the peak absorption coefficients were measured as 6.4 cm$^{-1}$, 12.2 cm$^{-1}$ and 16.3 cm$^{-1}$, respectively. However, this absorption peak is very sharp, with a full width at half maximum (FWHM) of 2.5 nm. There is also a broad absorption band at 940 nm that has double peaks centered at 935.6 nm and 939.6 nm, respectively. The two peaks have nearly the same absorption coefficient. The FWHM of this broad absorption band is 21.6 nm, which favors direct diode pumping with the InGaAs laser diodes. The absorption cross-section at 939 nm and 968 nm absorption peaks is calculated to be $0.73 \times 10^{-20}$ cm$^2$ and $0.91 \times 10^{-20}$ cm$^2$, respectively. In addition, an absorption peak at 1030 nm is also observable on the spectrum. The above measured results coincide with those of the previous reports on Yb:LuAG single crystals [18,19].

![Figure 2](image2)
Figure 3 displays the emission cross-section spectrum and fluorescent decay curve (inset) for a 5.0 at.% Yb:LuAG ceramic sample. The main emission peak at the 1030 nm wavelength has a FWHM of 6.8 nm. The emission cross-section at the peak is calculated to be $2.7 \times 10^{-20}$ cm$^2$ using the Füchtbauer–Ladenburg equation. The result is comparable with that of the Yb:LuAG single crystals reported previously [18]. The value is almost 30% higher than that of the Yb:YAG ($2.0 \times 10^{-20}$ cm$^2$). Laser gain media with a larger emission cross section will have a lower lasing threshold. The fluorescent lifetime of the doped ions in the sample was also measured, and a single exponential decay curve was observed. It gives a lifetime value of 0.96 ms. The lifetime value for the more highly doped samples were also measured. The value for 10% 15% 20% doping was measured to be 1.17ms, 1.21ms and 1.16ms, respectively. The value was slightly longer than the previously reported 10% Yb:LuAG ceramics of 1.07ms in Ref 23. The longer value is partially due to the radiation trapping effect of Yb$^{3+}$. 
The gain cross-section of the Yb:LuAG ceramics was then determined under different population inversion ratio $\beta$, which equals to $N_2/N_0$, where $N_2$ is the inversion population number and $N_0$ is the total population number. The gain cross-section $\sigma_g(\lambda)$ was calculated by: $\sigma_g(\lambda) = \beta \times \sigma_e(\lambda) - (1-\beta) \times \sigma_a(\lambda)$, where $\sigma_e(\lambda)$ and $\sigma_a(\lambda)$ are the emission and absorption cross-section, respectively. The gain cross-section of the Yb:LuAG ceramics under different $\beta$ was plotted in Fig. 4.

The laser performance of the 5.0 at % Yb:LuAG ceramic sample was investigated and was shown in Fig. 5. The Yb:LuAG ceramic sample was cut into $3 \times 3 \times 3$ mm$^3$ cubic and high
transmission coated on both its surfaces at the pump and lasing wavelengths. The sample was end-pumped by a fiber-coupled laser diode (Limo GmbH, LIMO25-F100-DL980) with a simple two-mirror cavity set-up. The delivery fiber of the pump laser had a core diameter of 100 µm and an N.A. of 0.22. The beam size in the sample was about 80 µm in radius.

Fig. 5. The CW laser performance of a 5.0 at.% Yb:LuAG ceramic sample under different output couplers.

CW operation at 1030 nm was successfully demonstrated with output couplers of different output couplings. Under an output coupler with \( T_{oc} = 50\% \), a maximum output power of 7.2 W was obtained at an absorbed pump power of 12.4 W, corresponding to a slope efficiency of 65%. We believe the result could be further improved by e.g. better cavity design, using ceramic samples with higher doping concentrations and shorter sample length, and better thermal management.

4. Conclusions

In conclusion, Yb:LuAG ceramics with various Yb-doping concentrations were successfully fabricated by using a solid-state reactive sintering method under vacuum condition. The fabricated samples have good optical quality. CW laser operation of the fabricated samples was demonstrated. It generated a maximum output power of 7.2 W with 65% slope efficiency. Nowadays, high power thin disk lasers have been developed and are one of the most promising solid state laser systems for many practical industry applications. The most often used gain medium for the thin-disk lasers is Yb:YAG crystal. Since the Yb:LuAG ceramics have 5-10% higher thermal conductivity and 30% higher emission cross section, it will be an attractive gain medium for the high average power thin disk lasers.

Acknowledgments

This research was supported by the National Research Foundation of Singapore under the contract NRF-G-CRP-2007-01.