

Fabrication, spectral and laser performance of 5 at.% Yb³⁺ doped (La_{0.10}Y_{0.90})₂O₃ transparent ceramic

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ABSTRACT

A 5 at.% Yb³⁺ doped (La_{0.10}Y_{0.90})₂O₃ transparent ceramic was fabricated with nano-powders and sintered in H₂ atmosphere. Spectroscopic properties and laser performance of Yb:(La_{0.10}Y_{0.90})₂O₃ ceramic were studied. The ceramic exhibits excellent spectroscopic properties, with broad absorption and emission bands, and its refractive index (*n*) is close to 2. The gain cross-section (σ_g) was calculated at different population inversion ratio (β) values. In addition, among Yb³⁺ doped YAG crystal, Y₂O₃ and (YLa)₂O₃ ceramic, (YLa)₂O₃ ceramic has the least pump intensity (I_{min}) of 1.25 KW cm⁻². Furthermore, a diode-pumped C–W ceramic laser output has been demonstrated at 1075 nm with a slope-efficiency of 60.2%.

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1. Introduction

The rare-earth-ions doped transparent ceramic, as promising laser materials, have been proved to be a good alternative to single-crystal laser materials [1,2]. Among them, Yb³⁺ doped transparent ceramic have attracted considerable attentions due to the development of the high-power and high-brightness InGaAs laser diodes [3]. Yb³⁺ ions have only two manifolds, ²F_{5/2} and ²F_{7/2}, with an energy gap of 10,000 cm⁻¹, which leads to a small quantum effect and avoids no intrinsic processes such as excited state absorption, cross relaxation, and concentration quenching. Furthermore, owing to perceptible electron–phonon interaction, Yb³⁺ doped laser materials have broad absorption in near IR benefits for laser diode (LD) pumping and broad luminescence band, which is favorable to the generation of ultra-short laser pulse [4].

Cubic Y₂O₃ has been investigated for many years as laser host material due to its excellent thermal conductivity (~13.6 W/km) [5], wide transparency range, high refractive index and low phonon energy. However, it is extremely difficult to grow high-quality and large-size Y₂O₃ single crystal using Czochralski method because of its high melting point (~2430 °C) and phase transformation at 2280 °C [6]. But it is much easier to fabricate Y₂O₃ transparent ceramic by the conventional ceramic processing because of the low sintering temperature, which is about 700 °C lower than its melting point by the nano-crystalline and non-press vacuum sintering technology [7]. In recent years, the slope efficiency of laser has reached 82.4% in Yb³⁺:Y₂O₃ ceramic [8], and the laser pulses

as short as 68 fs have also been reported in Yb³⁺ doped Y₂O₃ transparent ceramic [9].

(Y_{1-x}La_x)₂O₃ transparent ceramic, a solid solution of Y₂O₃ and La₂O₃, has cubic glass-like structure [10], so it has broader Yb³⁺ absorption and emission bandwidths compare to that of Y₂O₃ [11]. In this paper, the fabrication and spectral properties of 5 at.% Yb³⁺ doped (La_{0.10}Y_{0.90})₂O₃ transparent ceramic were investigated. In addition, laser properties of this material were also evaluated.

2. Experiment

High purity Yb³⁺ doped (La_{0.10}Y_{0.90})₂O₃ commercial nanopowders (10–15 nm) were used to fabricate ceramic (Rare-Chem. Hi-Tech Co., Ltd., China), and the Yb³⁺ concentration was 5 at.%. After calcined at 1200 °C for 5–10 h, the size of powders grows to 100–200 nm in order to get more compact green body, then the powders were ball milled in anhydrous alcohol for 5 h, dried, sieved, and pressed into disks with 15-mm-diameter and 5-mm-thickness. Finally disks were isostatically pressed at 200 MPa and sintered at 1680 °C for 45 h in H₂ atmosphere without pressure.

The sintered specimen was double polished for optical test, spectral analysis and laser experiment. The microstructure was observed with optical microscope (BX60, OLYPMUS, Japan). The refractive index was measured by spectroscopic ellipsometry (UVI/460-VIS-AGAS, Jobin Yvon S.A, France). The absorption spectrum was measured with a spectrophotometer using Xe light as pump source (Model V-570, JASCO, Japan). The fluorescence spectrum excited with 940 nm Xe lamp was measured with a

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spectrofluorimeter (Fluorolog-3, Jobin Yvon Spex, France). All the spectroscopic analyses were made at room temperature.

3. Results and discussion

Fig. 1 is the photo of Yb:(La_{0.10}Y_{0.90})₂O₃ transparent ceramic. All the specimens have high transparency. As shown in Fig. 2, the highest in-line transmittance is over 80%, which is close to its theoretical value. The ceramic refractive index (*n*) was decreased slightly with the increase of wavelengths as shown in Fig. 3, and close to 2 in the wavelength of ~1 μm, which is bigger than that of Y₂O₃ transparent ceramic (~1.8), therefore, the refractive index can be enhanced by adding La₂O₃. Fig. 4 shows the microstructure of Yb:(La_{0.10}Y_{0.90})₂O₃ transparent ceramic. There are almost no pores in or between the grain boundaries, and the grain size is between 30 and 50 μm.

Fig. 5 is the absorption and emission spectra at room temperature. There are three main absorption peaks centered at 903, 947, and 973 nm, and the full width at half maximum (FWHM) are 9.0, 15.5 and 7.8 nm, respectively. The strongest emission band corresponding to ²F_{7/2} → ²F_{5/2} transitions is centered at 1032 nm with FWHM of 15.7 nm, which covers wavelength range from 1000 to 1050 nm; the other main emission peak is located at 1074 nm with the FWHM of 19.9 nm.

Absorption cross-section (σ_{abs}) of Yb³⁺ can be determined by the following expression:

$$\sigma_{abs} = \frac{2.303 \log(I_0/I)}{L \cdot N} \quad (1)$$

where $\log(I_0/I)$ is optical density, *L* is thickness of the sample, and *N* is concentration of active ions in unit volume. The calculated absorption cross sections at 947 and 973 nm are $0.551 \times 10^{-20} \text{ cm}^2$ and $0.903 \times 10^{-20} \text{ cm}^2$, respectively.

The emission cross-sections (σ_{em}) of Yb³⁺ ion is determined by Fuchtbauer–Ladenburg (F–L) formula:

$$\sigma_{em}(\lambda) = \frac{1}{8\pi n^2 c} \frac{1}{\tau_{rad}} \frac{\lambda^5 I(\lambda)}{\int \lambda I(\lambda) d\lambda} \quad (2)$$

where τ_{rad} is the radiation lifetime, *c* is the light velocity in vacuum, *n* is the refractive index, and *I*(λ) is the emission intensity at wavelength λ . The calculated emission cross sections are: $\sigma_{em}(1032 \text{ nm}) = 0.896 \times 10^{-20} \text{ cm}^2$, $\sigma_{em}(1074 \text{ nm}) = 0.384 \times 10^{-20} \text{ cm}^2$, respectively.

Based on the absorption and emission cross-sections, the gain cross-section (σ_g) usually used to evaluate the laser properties of

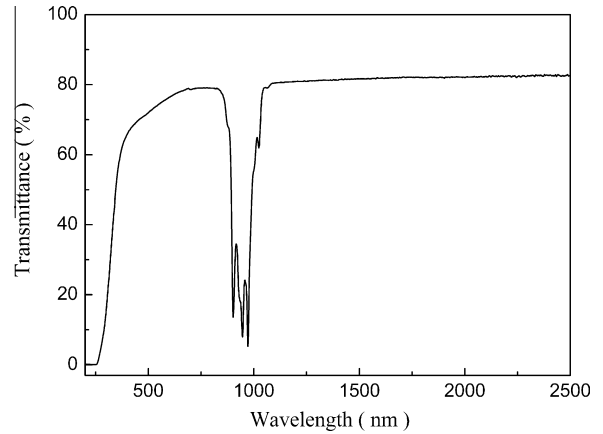


Fig. 2. In-line optical transmittance of Yb:(La_{0.10}Y_{0.90})₂O₃ transparent ceramic.

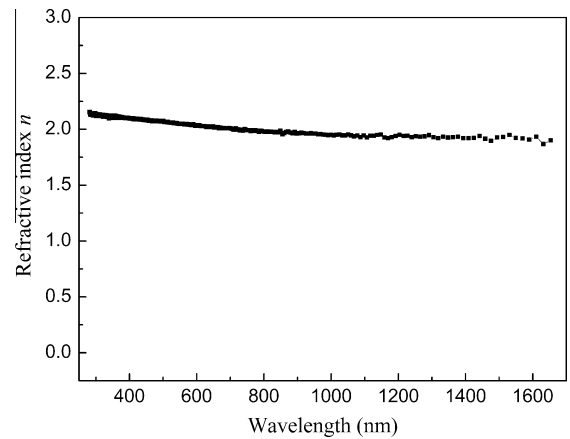


Fig. 3. Refractive index curve of Yb:(La_{0.10}Y_{0.90})₂O₃ transparent ceramic.

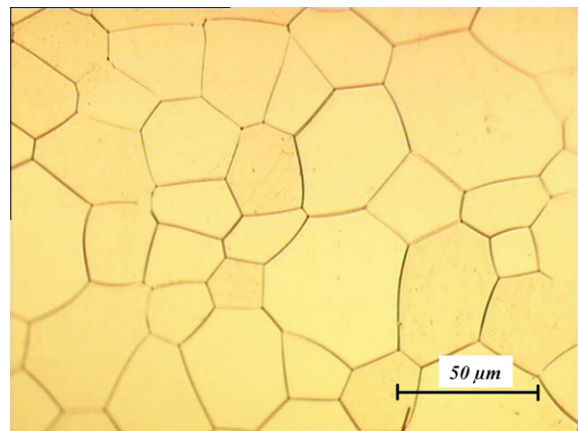


Fig. 4. Microscopic photograph of Yb:(La_{0.10}Y_{0.90})₂O₃ transparent ceramic.



Fig. 1. Photo of Yb:(La_{0.10}Y_{0.90})₂O₃ transparent ceramic.

materials for ²F_{5/2} → ²F_{7/2} transitions can be estimated by the following formula [12]:

$$\sigma_g = \beta \sigma_{em} - (1 - \beta) \sigma_{abs} \quad (3)$$

where β stands for Yb³⁺ ions population inversion ratio of the laser upper level.

Fig. 6 shows the relationships between the calculated gain cross-sections with different β values. When $\beta = 0.25$, the gain cross-section becomes positive at 1002 nm; the wavelength

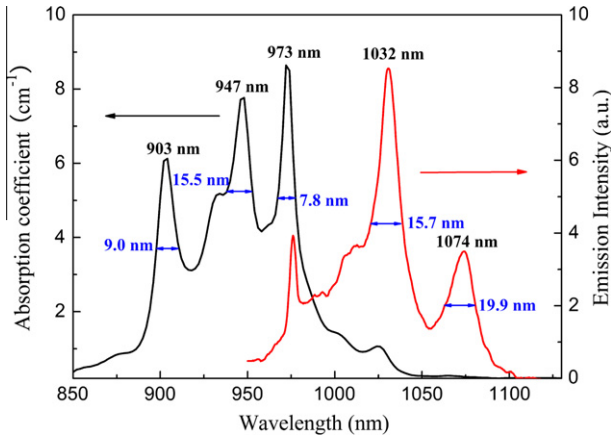


Fig. 5. Absorption and emission spectra of Yb:(La_{0.10}Y_{0.90})₂O₃ transparent ceramic.

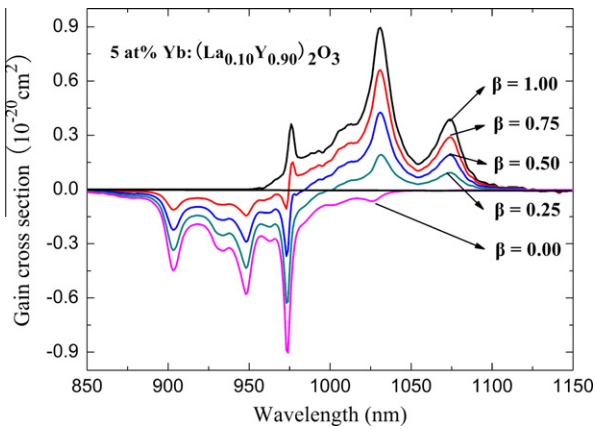


Fig. 6. Gain cross section of Yb:(La_{0.10}Y_{0.90})₂O₃ ceramic with different β values.

reaches 985 nm for $\beta = 0.5$; when $\beta = 0.75$, the gain curve indicates a positive gain from 975 to 1150 nm; and the emission cross-section spectra correspond to $\beta = 1$. It must be emphasized that the broad gain spectrum is favorable to achieve the ultra-short pulses laser, such as the femtosecond pulse laser.

Three laser parameters of the pump saturation intensity (I_{sat}), the minimum fraction (β_{min}), and the minimum pump intensity (I_{min}), are often used to evaluated whether the host materials is beneficial to realize laser output.

The pump saturation intensity (I_{sat}) is determined in the following equation:

$$I_{sat} = h\nu / (\sigma_{abs}\tau_{em}) \quad (4)$$

where h is Planck constant; ν is optical frequency; σ_{abs} is the absorption cross-section; τ_{em} is the emission lifetime.

The minimum fraction (β_{min}) is simply derived by:

$$\beta_{min} = \frac{\sigma_{abs}(\lambda_{ext})}{\sigma_{ext}(\lambda_{ext}) + \sigma_{abs}(\lambda_{ext})} \quad (5)$$

where $\sigma_{abs}(\lambda_{ext})$ and $\sigma_{ext}(\lambda_{ext})$ are the absorption and emission cross-sections at laser wavelength λ_{ext} , respectively.

Table 1
Laser parameters comparison of Yb³⁺ doped YAG crystal, Y₂O₃ and (La_{0.10}Y_{0.90})₂O₃ ceramics.

Host	λ_{pump} (nm)	I_{sat} (kW cm ⁻²)	I_{min} (kW cm ⁻²)	λ_{ext} (nm)	β_{min}	σ_{ext} (10 ⁻²¹ cm ²)	Refs.
YAG	942	27.9	1.53	1031	0.055	20.3	[13]
Y ₂ O ₃	977	29.3	1.43	1030	0.049	9.50	[14]
LaYO ₃	976	32.2	1.25	1032	0.039	8.96	This work

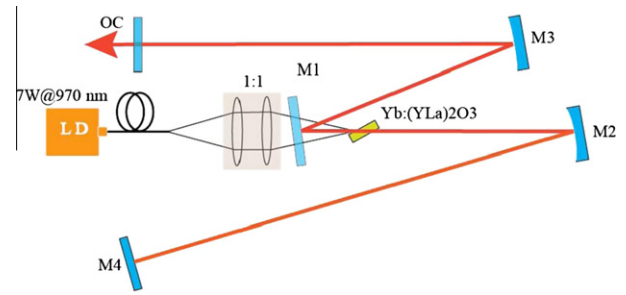


Fig. 7. Schematic of the laser experimental setup (CW operation).

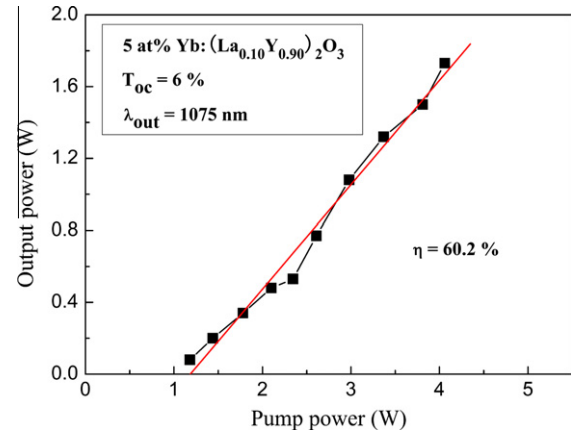


Fig. 8. Output power versus absorbed power of Yb:(La_{0.10}Y_{0.90})₂O₃ ceramic laser.

The minimum pump intensity (I_{min}) is given by:

$$I_{min} = \beta_{min} I_{sat} \quad (6)$$

Laser parameters of Yb³⁺ doped YAG crystal, Y₂O₃ and (La_{0.10}Y_{0.90})₂O₃ ceramic are listed in Table 1. YAG crystal has the least saturation pump intensities I_{sat} of 27.9 kW cm⁻², and (La_{0.10}Y_{0.90})₂O₃ ceramic has the least pump intensity I_{min} and β_{min} of 1.25 kW cm⁻² and 0.039, respectively, it can predict that the Yb:(La_{0.10}Y_{0.90})₂O₃ ceramic has lower threshold pump power, general lower fraction β_{min} means that it is easier for Yb³⁺ ions to reach population inversion in Yb:(La_{0.10}Y_{0.90})₂O₃ transparent ceramic.

An uncoated Yb:(La_{0.10}Y_{0.90})₂O₃ ceramic was used for lasing experiment under LD end-pumping. The ceramic plate's dimension is 3 × 5 × 2 mm³. The pump light at 976 nm from a fiber coupled LD bar was focused into the ceramic by two coupling lenses, and the focused pump beam had a diameter of 50 μ m. A Z-fold cavity shown in Fig. 7 was employed for CW experiment [15], plane dichroic mirror with high transmission at 976 nm and high reflection at 1020–1100 nm was applied. In addition, plane mirror with transmissions 6% was used as output couplers (OCs).

The laser output power versus the absorbed power of the ceramic laser is shown in Fig. 8. The threshold is about 1.2 W with a slope efficiency of 60.2%. With a pump power of 4 W, a maximum output power of 1.73 W was obtained at the wavelength 1075 nm, and no saturation was reached, which indicates that a higher output power will be obtained with a higher pumping power. So it is

believed that more high-power and efficient lasers will be demonstrated by optimizing the thickness of gain medium coated with anti-reflection coatings, Yb³⁺ concentration, and laser cavity.

4. Conclusions

High optical quality 5 at.% Yb:(La_{0.10}Y_{0.90})₂O₃ transparent ceramic was successfully fabricated with nanopowders. The ceramic has a high refractive index (n) of around two. The main absorption peaks at 947 and 973 nm have a FWHM of 15.5 and 7.8 nm, and the main emission peaks at 1032 and 1074 nm have a FWHM of 15.7 and 19.9 nm, respectively. The gain cross-sections for different population inversion ratio (β) values were calculated. The least pump intensity (I_{min}) of ceramic is 1.25 KW cm⁻². A maximum output power of 1.73 W with a slope efficiency of 60.2% was demonstrated in Yb:(La_{0.10}Y_{0.90})₂O₃ ceramic laser.

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References

- [1] H. Yagi, T. Yanagitani, K. Takaichi, *Opt. Mater.* 29 (2007) 1258.
- [2] Q.H. Yang, S.Z. Lu, B. Zhang, H.J. Zhang, J. Zhou, Z.J. Yuan, Y.F. Qi, Q.H. Lou, *Opt. Mater.* 33 (2011) 692.
- [3] J. Sanghera, W. Kim, C. Baker, G. Villalobos, J. Frantz, B. Shaw, A. Lutz, B. Sadowski, R. Miklos, *Opt. Mater.* 33 (2011) 670.
- [4] A. Shirakawa, K. Takaichi, H. Yagi, M. Tanisho, J.F. Bisson, J. Lu, K. Ueda, T. Yanagitani, A.A. Kaminskii, *Laser Phys.* 11 (2004) 1375.
- [5] U. Griebner, V. Petrov, K. Petermann, V. Peters, *Opt. Exp.* 12 (2004) 3125.
- [6] L. Fornasiero, E. Mix, V. Peters, K. Petermann, G. Huber, *Cryst. Res. Technol.* 34 (1999) 255.
- [7] K. Takaichi, H. Yagi, J. Lu, J. Bisson, A. Shirakawa, K. Ueda, T. Yanagitani, K. Takuma, A. Kaminskii, *Appl. Phys. Lett.* 84 (2004) 317.
- [8] J. Kong, D.Y. Tang, C.C. Chan, J. Lu, K. Ueda, H. Yagi, T. Yanagitani, *Opt. Lett.* 32 (2007) 247.
- [9] M. Tokurakawa, A. Shirakawa, K. Ueda, H. Yagi, M. Noriyuki, T. Yanagitani, A.A. Kaminskii, *Opt. Express* 17 (2009) 3353.
- [10] M. Yoshimura, X.Z. Rong, *J. Mater. Sci. Lett.* 16 (1997) 1961.
- [11] Q.H. Yang, C.G. Dou, J. Ding, X.M. Hu, J. Xu, *Appl. Phys. Lett.* 91 (2007) 111918.
- [12] I. Razdobreev, L. Bigot, V. Pureur, A. Favre, G. Bouwmans, M. Douay, *Appl. Phys. Lett.* 90 (2007) 031103.
- [13] L.D. DeLoach, S.A. Payne, L.L. Chase, L.K. Smith, W.L. Kaway, W.F. Krupke, *IEEE J. Quantum Electron.* 29 (1993) 1179.
- [14] J. Kong, D.Y. Tang, J. Lu, K. Ueda, *Appl. Phys. B* 79 (2004) 449.
- [15] Y.D. Zhang, Z.Y. Wei, Z.L. Wang, Z.G. Zhang, H.J. Zhang, Q.H. Yang, *IQEC/CLEO Pacific Rim* 978 (2011) 1844.