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Optimization of Bi_2O_3–B_2O_3-based glass phosphor co-doped with Yb^{3+} and Nd^{3+} for optical coherence tomography light source

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Abstract We have optimized Yb_2O_3 concentration, Nd_2O_3 concentration, and thickness of a Yb^{3+}, Nd^{3+} co-doped Bi_2O_3–B_2O_3-based glass phosphor to achieve a higher output power with a Gaussian-like shaped luminescence for an optical coherence tomography (OCT) light source. A simple model that includes absorption of excitation light, absorption of luminescent light, and luminescence efficiency was used for estimation of the output power. We have estimated from the model that the highest output power was achieved at Yb_2O_3 concentration of 3%, Nd_2O_3 concentration of 0.5%, and the thickness of 1.8 mm. This estimation showed a good agreement with the experimental results. For the optimized glass phosphor, we have observed Gaussian-like shaped luminescence, which is important for the OCT light source.

1 Introduction

Wideband light sources are used in a fiber gyroscope [1, 2] and optical coherence tomography (OCT), that is, a recently developed cross-sectional imaging technique for biological tissues [3]. Since OCT is based on a Michelson interferometer, a low-coherence wideband light source has several advantages for a high depth-resolution. The depth-resolution Δz, which is equal to the coherence length in the Michelson interferometer, is calculated from the equation:

$$\Delta z = \frac{2\ln 2}{\pi}\frac{\lambda_c^2}{\Delta\lambda}, \tag{1}$$

where λ_c is the central wavelength and $\Delta\lambda$ is the full width at half maximum (FWHM) of a Gaussian-shaped spectrum. It is obvious from this equation that a shorter central wavelength results in a higher depth-resolution. However, the central wavelength is limited to a near-infrared region in the present applications, leading to penetration depths in biological tissues much larger than that at other wavelengths [3]. A water absorption profile has a local minimum at $\lambda \sim 1060$ nm. In addition to it, a recent study has shown that imaging biological tissues at a central wavelength around 1000 nm yields minimal OCT axial resolution degradation due to water dispersion [4]. Therefore, the central wavelength λ_c should be near 1000 nm with a wider spectral width for the higher depth-resolution.

Super luminescent diodes (SLDs) and light emitting diodes (LEDs) in the near-infrared region are usually used in OCT. The spectral width of SLDs and LEDs is 50 nm at the maximum, corresponding to the coherence length of around 10 μm. These light sources are simple and not very expensive. However, this coherence length of around 10 μm is insufficient for several medical applications. Recently, using a super continuum laser, the coherence length of less than 10 μm was achieved [3]. However, this light source is complex, quite expensive, and difficult to use in practical applications in a hospital setting. Zhang et al. have reported a method for a synthesis of several LEDs to shorten the coherence length [5]. However, such synthesis of several LEDs and control of light intensity of individual LEDs are complicated for practical uses.

Therefore, we propose a novel near-infrared light source for OCT by combining a near-infrared phosphor and an LED in one package. To realize this light source, we synthesized the phosphor that emits at around 1000 nm using Bi_2O_3–B_2O_3-based glasses doped with Yb^{3+} and Nd^{3+} [6, 7]. And we reported that a high power (over 1 mW) near-infrared

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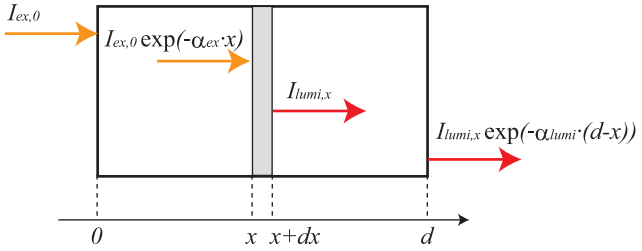

Fig. 1 Schematic drawing of the calculation model. The excitation light of $I_{ex,0}$ comes from left-side of the glass phosphor having the thickness d. In this model, the light travels only to the $+x$ direction

light source was successfully achieved by using this glass phosphor and a high power amber LED [8]. In this paper, we report details of optimization of Yb_2O_3 concentration, Nd_2O_3 concentration, and thickness of the Yb^{3+}, Nd^{3+} co-doped Bi_2O_3–B_2O_3-based glass phosphor to achieve a higher output power with Gaussian-like shaped luminescence.

2 Sample preparation

The glass phosphor used in this study was synthesized by melt-quenching. Powders of Yb_2O_3, Nd_2O_3, Bi_2O_3, H_3BO_3, and Sb_2O_3 were mixed with nominal molar compositions of $x\,Yb_2O_3$–$y Nd_2O_3$–$(99–x–y)(50Bi_2O_3–50B_2O_3)$–$1.0Sb_2O_3$. The mixed powders in an Al_2O_3 crucible were melted at 1250 °C in an electric furnace. Sb_2O_3 was used to suppress reduction of Bi^{3+}. After keeping the mixture for 10 min, the molten liquid was poured between two stainless steel mould plates that were kept at room temperature for a formation of the glass.

3 Calculation of output intensity

To calculate the output intensity, we consider the glass phosphor having the thickness d. It is assumed that a light travels only to $+x$ direction in the glass phosphor. As shown in Fig. 1, an excitation light intensity $I_{ex,x}$ is generally given by

$$I_{\mathrm{ex},x} = I_{\mathrm{ex},0}\exp(-\alpha_{\mathrm{ex}}\cdot x), \tag{2}$$

where α_{ex} is an absorption coefficient of the excitation light. The output intensity of a luminescent light from a position x is described as

$$I_{\mathrm{out},x} = I_{\mathrm{lumi},x}\exp\big(-\alpha_{\mathrm{lumi}}\cdot(d-x)\big), \tag{3}$$

where α_{lumi} is the absorption coefficient of the luminescent light and $I_{\mathrm{lumi},x}$ is the intensity of the luminescent light at the position x.

The absorbed excitation light in an infinitesimal layer of a thickness dx is described as

$$\begin{aligned} &I_{\mathrm{ex},x} - I_{\mathrm{ex},x+dx} \\ &= I_{\mathrm{ex},0}\exp(-\alpha_{\mathrm{ex}}\cdot x) - I_{\mathrm{ex},0}\exp\big(-\alpha_{\mathrm{ex}}\cdot(x+dx)\big) \\ &= I_{\mathrm{ex},0}\exp(-\alpha_{\mathrm{ex}}\cdot x)\cdot\big(1-\exp(-\alpha_{\mathrm{ex}}\cdot dx)\big) \\ &\approx I_{\mathrm{ex},0}\alpha_{\mathrm{ex}}\exp(-\alpha_{\mathrm{ex}}\cdot x)\,dx. \end{aligned} \tag{4}$$

Therefore, $I_{\mathrm{lumi},x}$ is calculated from the equation

$$I_{\mathrm{lumi},x} = \eta I_{\mathrm{ex},0}\alpha_{\mathrm{ex}}\exp(-\alpha_{\mathrm{ex}}\cdot x)\,dx, \tag{5}$$

where η is luminescence efficiency. By using (3) and (5), the total output intensity I_{out} is described as

$$\begin{aligned} I_{\mathrm{out}} &= \int_0^d I_{\mathrm{out},x}\,dx \\ &= \eta I_{\mathrm{ex},0}\alpha_{\mathrm{ex}}\exp(-\alpha_{\mathrm{lumi}}\cdot d) \\ &\quad\times\int_0^d \exp\big((\alpha_{\mathrm{lumi}}-\alpha_{\mathrm{ex}})\cdot x\big)\,dx \\ &= \eta I_{\mathrm{ex},0}\alpha_{\mathrm{ex}}\exp(-\alpha_{\mathrm{lumi}}\cdot d) \\ &\quad\times\big(\exp\big((\alpha_{\mathrm{lumi}}-\alpha_{\mathrm{ex}})\cdot d\big)-1\big)/(\alpha_{\mathrm{lumi}}-\alpha_{\mathrm{ex}}). \end{aligned} \tag{6}$$

Therefore, to calculate I_{out}, we have to estimate α_{ex}, α_{lumi}, and η.

Firstly, we estimate α_{ex}. Figure 2 shows an excitation and luminescent process, which is considered from a photoluminescence excitation (PLE) and photoluminescence (PL) measurements of the present glass phosphor [6–8]. Luminescence of Nd^{3+} should be caused mainly by the excitation of the host glass and that of Nd^{3+}. Luminescence of Yb^{3+} should be caused mainly by the excitation of the host glass and the energy transfer from Nd^{3+}. Since the amber LED ($\lambda_c = 590$ nm) is used for the excitation light source in this study, we measure the absorption coefficient at 590 nm. From Fig. 2, it is obvious that α_{ex} consists of the absorption by Nd^{3+} and host glass. We fix the Yb_2O_3 concentration at 1 mol%. We measure the absorption coefficient at the Nd_2O_3 concentration to be equal to or lower than 4 mol%, since a concentration quenching occurs over 5 mol% [7]. Figure 3 shows dependence of the absorption coefficient on the Nd_2O_3 concentration at 590 nm. A linear dependence of the absorption coefficient is clearly observed. From Fig. 3, we estimate α_{ex} as

$$\alpha_{\mathrm{ex}} = 0.38 + 0.19\cdot C_{\mathrm{Nd_2O_3}}, \tag{7}$$

where $C_{\mathrm{Nd_2O_3}}$ is the Nd_2O_3 concentration.

Secondly, we focus on α_{lumi}. From our previous study, α_{lumi} should consist of the absorption by Yb^{3+} and host glass [6]. However, in our previous study, Yb^{3+} showed a wideband absorption spectrum [6]. Therefore, in this study,

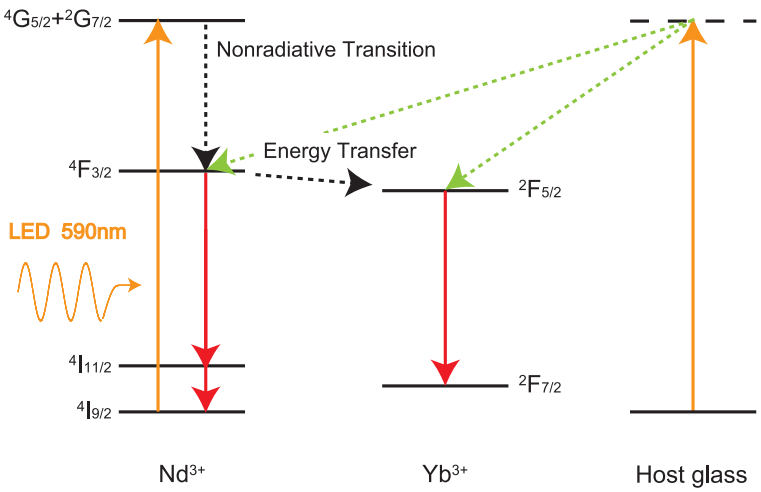

Fig. 2 Schematic drawing of the absorption and the luminescent processes. The excitation light ($\lambda_c = 590$ nm) is absorbed by Nd^{3+} ions and the host glass. Luminescence of Nd^{3+} should be excited mainly by the host glass and Nd^{3+}. The luminescence of Yb^{3+} should be excited mainly by the host glass and energy transfer from Nd^{3+}

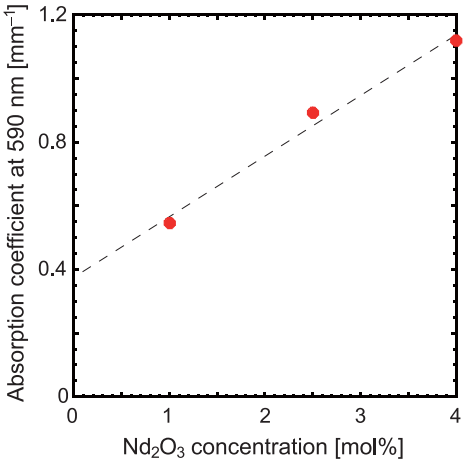

Fig. 3 Dependence of the absorption coefficient on Nd_2O_3 concentration at 590 nm. Linear dependence of the absorption coefficient is clearly observed

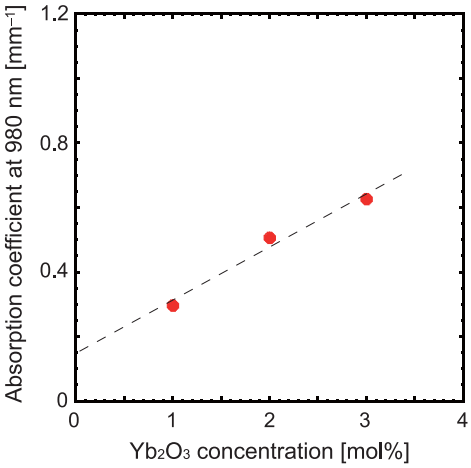

Fig. 4 Dependence of the absorption coefficient on Yb_2O_3 concentration at 980 nm. Linear dependence of the absorption coefficient is clearly observed

an absorption peak around 980 nm is measured for estimation of α_{lumi}. We fix the Nd_2O_3 concentration at 4 mol%. We measure the absorption coefficient at the Yb_2O_3 concentration equal to or lower than 3 mol%, since a concentration quenching occurs over 4 mol% [7]. Figure 4 shows a dependence of the absorption coefficient on Yb_2O_3 concentration at 980 nm. A linear dependence of the absorption coefficient is clearly observed. From Fig. 4, α_{lumi} is estimated as

$$\alpha_{\mathrm{lim}\,i} = 0.15 + 0.17 \cdot C_{\mathrm{Yb_2O_3}}, \tag{8}$$

where $C_{\mathrm{Yb_2O_3}}$ is the Yb_2O_3 concentration.

Finally, we estimate η. As mentioned above, luminescence of the present glass phosphor should consist mainly of luminescence of Nd^{3+} excited by the host glass, that of Nd^{3+} excited by Nd^{3+}, that of Yb^{3+} excited by the host glass, and that of Yb^{3+} by the transferred energy from Nd^{3+}. Therefore, we assume that η is described by

$$\eta = A_1 \cdot C_{\mathrm{Nd_2O_3}} + A_2 \cdot C_{\mathrm{Yb_2O_3}} + A_3 \cdot C_{\mathrm{Nd_2O_3}} \cdot C_{\mathrm{Yb_2O_3}}. \tag{9}$$

In (9), the first term represents luminescence of Nd^{3+} excited by the host glass and Nd^{3+}. The second term represents luminescence of Yb^{3+} excited by the host glass. And the third term represents luminescence of Yb^{3+} by the transferred energy from Nd^{3+}. Since it is very complex to consider each processes, we estimate η by measuring $I_{\mathrm{ex},0}$, I_{out}, α_{ex}, α_{lumi}, and d, and substitute these values into (6). The estimated η of several glass phosphors are shown in Table 1. From Table 1 and (6), we roughly estimate η as

$$\eta = 0.013 \cdot C_{\mathrm{Nd_2O_3}} + 0.058 \cdot C_{\mathrm{Yb_2O_3}} - 0.015 \cdot C_{\mathrm{Nd_2O_3}} \cdot C_{\mathrm{Yb_2O_3}}. \tag{10}$$

Using (6), (7), (8), and (10), we can calculate the output intensity of the luminescent light. From calculations of the

output power, we estimate that the highest output power is achieved at the Yb_2O_3 concentration of 3%, Nd_2O_3 concentration of 0.5%, and the thickness of 1.8 mm.

4 Results and discussion

Figures 5(a), 5(b), and 5(c) show the calculated $I_{out}/I_{ex,0}$ (dashed line) and the experimental $I_{out}/I_{ex,0}$ (circle) as a function of the Yb_2O_3 concentration, the Nd_2O_3 concentration, and the thickness of the glass phosphor, respectively. In Fig. 5(a), it is clearly observed that the calculated $I_{out}/I_{ex,0}$ increases with increasing Yb_2O_3 concentration. On the other hand, the calculated $I_{out}/I_{ex,0}$ decreases beyond the Nd_2O_3 concentration of 0.5 mol% as shown in Fig. 5(b), and beyond the thickness of 1.8 mm as show in Fig. 5(c). The value of the highest $I_{out}/I_{ex,0}$ in the calculation results shows good agreement with that in the experimental results. The dependence of $I_{out}/I_{ex,0}$ on the thickness indicates that the balance of luminescence and absorption of near-infrared light is important in this glass phosphor. Though there are differences between calculation and experiment in absolute values, dependence of Yb_2O_3 concentration and the thickness on $I_{out}/I_{ex,0}$ are similar. These differences in absolute values should be due to the model used, which assumes the light travels only to $+x$ direction, and neglects reflection and scattering. On the other hand, the calculated dependence of Nd_2O_3 concentration on $I_{out}/I_{ex,0}$ disagrees with the experimental dependence. We consider that this disagreement is due to rough estimation of η shown in (9). Especially, interaction between Nd^{3+} and Yb^{3+} should not be so simple as a multiplication of $C_{Nd_2O_3}$ and $C_{Yb_2O_3}$ [9]. The detailed analysis of the excitation rate of Nd^{3+}, the energy transfer rate from Nd^{3+} to Yb^{3+}, the energy back transfer rate from Nd^{3+} to Yb^{3+}, and the luminescence rate of Yb^{3+} are needed to clarify the dependence of Nd_2O_3 concentration and Yb_2O_3 concentration on luminescence efficiency.

Figure 6 shows the PL spectrum of $3.1Yb_2O_3$–$0.5Nd_2O_3$–$47.6Bi_2O_3$–$47.8B_2O_3$–$1.0Sb_2O_3$ with a thickness of 2 mm. A Gaussian-like shaped spectrum is observed with a peak at

Table 1 Estimated luminescence efficiency of several glass phosphors

$C_{Yb_2O_3}$ [mol%]	$C_{Nd_2O_3}$ [mol%]	η [%]
1	1	5.6
1	2.5	5.1
1	4	4.9
2	4	4.8
3	4	5.8

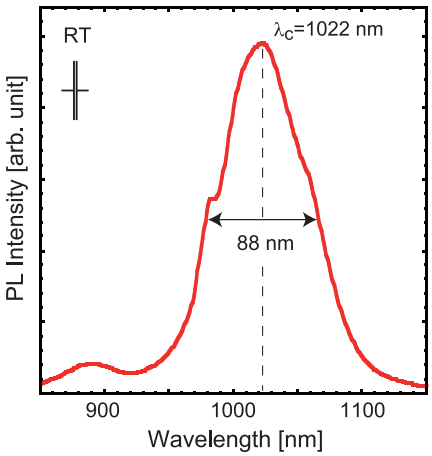

Fig. 6 The PL spectrum of $3.1Yb_2O_3$–$0.5Nd_2O_3$–$47.6Bi_2O_3$–$47.8B_2O_3$–$1.0Sb_2O_3$ with the thickness of 2 mm. A Gaussian-like shaped spectrum is observed with the peak at 1022 nm and the FWHM as wide as 88 nm

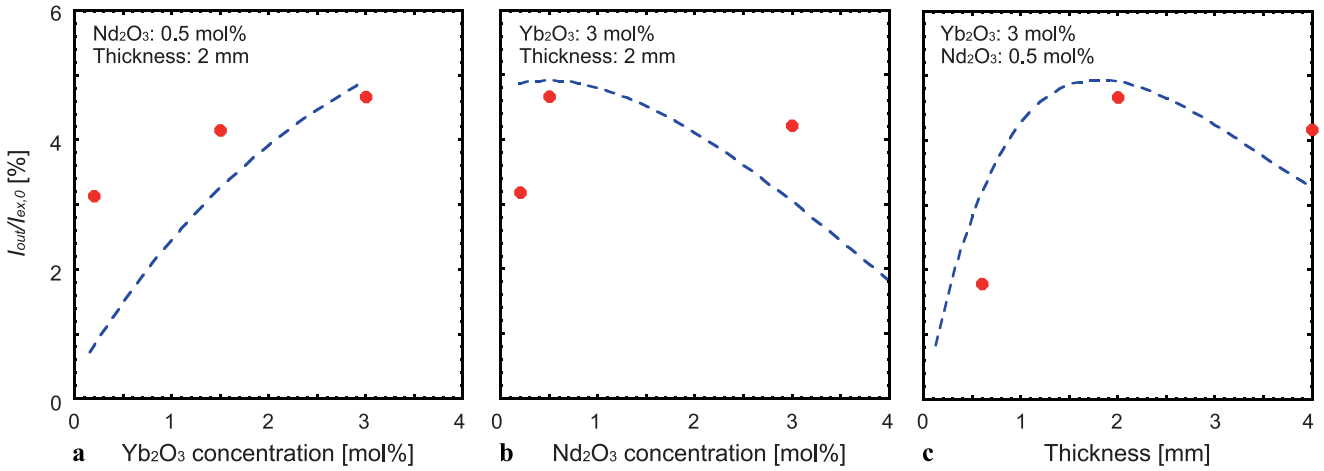

Fig. 5 The calculated $I_{out}/I_{ex,0}$ (*dashed line*) and the experimental $I_{out}/I_{ex,0}$ (*circle*) as functions of (**a**) Yb_2O_3 concentration, (**b**) Nd_2O_3 concentration, and (**c**) the thickness of the glass phosphor, respectively. The value of the highest $I_{out}/I_{ex,0}$ in the calculation results shows good agreement with that of the experimental results

1022 nm and a FWHM as wide as 88 nm. The asymmetry of spectrum is caused by the luminescence shape of Yb^{3+} (around 980 nm) and the luminescence of Nd^{3+} (around 1060 nm) [6]. The small peak around 900 nm is the luminescence of Nd^{3+} [6]. The coherence length is estimated to be 5.2 μm. This coherence length should improve the depth-resolution of OCT by twice compare with that of conventional SLDs and LEDs. Therefore, our near-infrared light source is indeed useful for the OCT light source. These results suggest that the simple model using this study is a useful tool to optimize concentration of rare-earth ions and the thickness of the glass phosphor.

5 Conclusion

We have optimized Yb_2O_3 concentration, Nd_2O_3 concentration, and the thickness of the glass phosphor to achieve the highest output power of the near-infrared light source using the glass phosphor and the LED. We estimated the output power by using a simple model including the absorption of the excitation light, the absorption of the luminescent light, and the luminescence efficiency. By measuring the absorption coefficient of the excitation light, the absorption coefficient of the luminescent light, and luminescence efficiency, we have calculated the output power. From the calculation results, we estimated that the highest output power is achieved at a Yb_2O_3 concentration of 3%, a Nd_2O_3 concentration of 0.5%, and a thickness of 1.8 mm. In the synthesized samples, the highest output power was achieved from $3.1Yb_2O_3$–$0.5Nd_2O_3$–$47.6Bi_2O_3$–$47.8B_2O_3$–$1.0Sb_2O_3$ with the thickness of 2 mm. We observed Gaussian-like shaped luminescence with the peak at 1022 nm and the FWHM as wide as 88 nm. These results indicate that the calculation model using this study is a useful tool for optimization of Yb_2O_3 concentration, Nd_2O_3 concentration, and the thickness of the Yb^{3+}, Nd^{3+} co-doped Bi_2O_3–B_2O_3-based glass phosphor.

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References

1. C.C. Cutler, S.A. Newton, H.J. Shaw, Opt. Lett. **5**, 488 (1980)
2. S. Blin, H.K. Kim, M.J.F. Digonnet, G.S. Kino, J. Lightwave Technol. **25**, 861 (2007)
3. A.F. Fercher, W. Drexler, C.K. Hitzenberger, T. Lasser, Rep. Prog. Phys. **66**, 239 (2003)
4. Y. Wang, J. Nelson, Z. Chen, B. Reiser, R. Chuck, R. Windeler, Opt. Express **11**, 1411 (2003)
5. Y. Zhang, M. Sato, N. Tanno, Opt. Lett. **26**, 205 (2001)
6. S. Fuchi, A. Sakano, Y. Takeda, Jpn. J. Appl. Phys. **47**, 7932 (2008)
7. S. Fuchi, A. Sakano, R. Mizutani, Y. Takeda, Eur. J. Glass Sci. Technol. A **50**, 319 (2009)
8. S. Fuchi, A. Sakano, R. Mizutani, Y. Takeda, Appl. Phys. Express **2**, 32102 (2009)
9. F. Batalioto, D.F. de Sousa, M.J.V. Bell, R. Lebullenger, A.C. Hernandes, L.A.O. Nunes, J. Non-Cryst. Solids **273**, 233 (2000)