



Fabrication of waveguides by fs-laser micromachining in Dy³⁺/Eu³⁺ doped barium borate glass with broad emission in the visible spectrum



S.N.C. Santos ^{*}, J.M.P. Almeida, G.F.B. Almeida, V.R. Mastelaro, C.R. Mendonca

São Carlos Institute of Physics, University of São Paulo, PO Box 369, 13560-970, São Carlos, SP, Brazil

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ABSTRACT

Boron-based glasses are relevant materials for optical applications on account its ability to hold large amount of oxide modifiers and rare-earth ions. The barium borates ones are particularly interesting due to its designed second and third-order optical nonlinearities. Despite the interest on barium borate glasses for linear and nonlinear optics, the fabrication of active elements for optical microcircuits are still scarce. In this paper, we demonstrate the fabrication of three dimensional waveguides in barium borate glass with broad emission in the visible spectrum. Such performance was achieved through the co-doping with Dy³⁺ and Eu³⁺ ions, which together provide broad emissions at blue, yellow and red, along with the glass processing with femtosecond laser pulses. Almost circular waveguides, with diameter of approximately 5 μm and 7.8 mm long, were inscribed into the bulk of the glass by direct laser writing, whose support single mode guiding at 632.8 nm with propagation loss of (3 ± 1) dB/cm. When coupling UV light, such waveguides display the typical emission of Dy³⁺ and Eu³⁺ ions with its additional guiding throughout the waveguide length. The broad band emission and its guiding in the visible spectrum reveal that the waveguides fabricated by femtosecond laser micromachining in co-doped barium borate glass are promising active elements for the development of optical micro-devices.

1. Introduction

Boron-based glasses are one of the major glass forming matrices, which present low production cost, thermal stability, wide range of transparency, ease incorporation of oxides modifier and rare-earth ions [1,2]. Therefore, the composition of boron-based glasses can be easily changed to achieve specific properties targeting different applications. For instance, the barium borate ones can be designed to have second harmonic generation by submitting it to crystallization or polling processes [3,4]. Besides its relevance for second-order optical nonlinearities, barium borate glasses can also exhibit important third-order nonlinear properties, reaching higher values when doped with rare-earth ions [5].

Over the years, the interest in exploring rare-earth doped or co-doped ions in glassy materials has increased. In particular, borate glasses have been extensively studied because of their high rare-earth ions solubility [6], which is interesting for the development of the photonic and optoelectronic devices [7–12]. Depending on the rare-earth ion, different functionalities can be accomplished; for example, the incorporation of Nd³⁺ ions in barium borate matrix was demonstrate to be promising for broadband laser amplification in the infrared region [13].

Also, energy applications were demonstrate in barium borate glasses co-doped with Eu³⁺–Tb³⁺, being efficient as photon downshifting cover glass for solar cells [14].

Among the lanthanide ions, dysprosium has unique luminescence properties. The glass-doped Dy³⁺ ion exhibits two dominant emission bands in the blue (470–500 nm) and yellow (560–600 nm) regions, with applications as radiation dosimetry measurement and solid state lasers [15,16]. Also, Eu³⁺ ion has been used as a dopant for glass matrix targeting at several applications, such as white OLEDs, LED and plasma display [17–19] due to its characteristic red emission at approximately 616 nm. The combination of emissions from Dy³⁺ and Eu³⁺ ions can result in white light generation, which is promising for photonic devices [20]. Although the co-doping of Dy³⁺ and Eu³⁺ is interesting for luminescent glasses, its incorporation in barium borate glasses for the development of optical devices is still scarce. At the same time, femtosecond laser micromachining has been shown as an important method for the fabrication of optical devices in transparent materials, being useful for processing glassy materials, allowing obtaining gain waveguides and amplifiers [21–24].

In this paper, considering the interests on luminescent borate glasses and the advantages of material processing with fs-lasers, we

^{*} Corresponding author.

E-mail addresses: ncs.sabrina@gmail.com (S.N.C. Santos), crmendon@ifsc.usp.br (C.R. Mendonca).

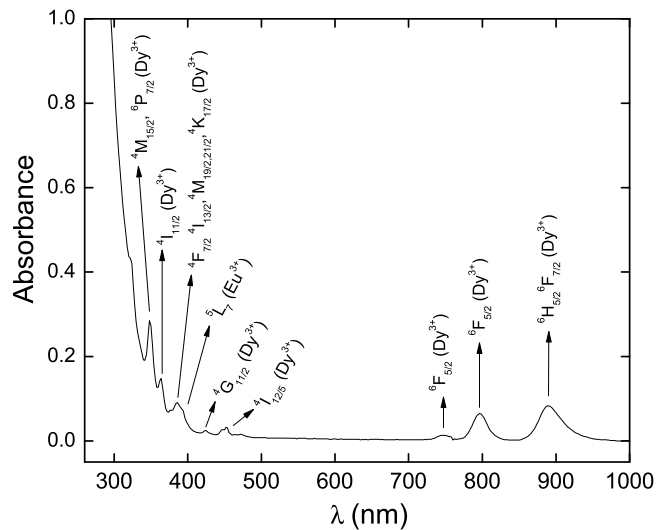


Fig. 1. Absorption spectrum of Dy^{3+} and Eu^{3+} co-doped B_2O_3 – BaO – SiO_2 glass with the assigned transitions.

demonstrate the fabrication of three dimensional optical waveguides in Dy^{3+} – Eu^{3+} co-doped barium borate glasses with broad band emission in the visible spectrum. The waveguides were fabricated by femtosecond laser micromachining with pulse energy of 32 nJ and 10 $\mu\text{m}/\text{s}$ of scan speed, which resulted in homogeneous, 7.8 mm-long waveguides with slight elliptical cross-section of approximately 5 μm . Besides the Gaussian distribution guiding profile, with overall losses of (7 ± 1) dB at 632.8 nm, including a propagation loss of (3 ± 1) dB/cm, such waveguides display the guiding of the Dy^{3+} and Eu^{3+} ions emission, configuring an important active optical element for white light generation.

2. Experimental

The glass sample composition studied here is based on a barium borate matrix doped with the rare-earth ions Dy^{3+} and Eu^{3+} . Such sample was prepared by the melt-quenching method employing a platinum crucible using an electric furnace and high purity (>99.99%) borate, barium and silicon oxides. Subsequently, Dy_2O_3 (0.1 mol%) and Eu_2O_3 (0.05 mol%) were incorporated into this matrix also by melt-quenching, as described in Ref. [25], yielding a glass sample with the chemical composition of $42.5\text{B}_2\text{O}_3$ – 42.5BaO – 15SiO_2 : $0.1\text{Dy}_2\text{O}_3$ – $0.05\text{Eu}_2\text{O}_3$ (mol%), here named as BBS-DyEu. Waveguides were written throughout the length of the sample by a Ti: Sapphire laser oscillator delivering 50-fs pulses, centered at 800 nm, with maximum energy of 100 nJ and operating at a repetition rate of 5 MHz. Femtosecond laser pulses were focused by a microscope objective (NA = 0.65) within the sample, while it was displaced by a translational xyz stage moved at 10 $\mu\text{m}/\text{s}$ in the plane perpendicular to the laser beam. The guiding properties of the produced waveguides were determined by using an objective-lens based coupling system; standard and UV-coated microscopy objectives (NA = 0.65) were used to coupled light from a He–Ne (632.8 nm) and a He–Cd (325 and 442 nm), respectively. The guided modes, including the guided fluorescence, were observed with the aid of a CCD camera. Waveguide losses were determined by measuring the input and output laser power, considering the transmission of all optical components in the system [26].

3. Results and discussions

The absorption spectrum of the sample, presented in Fig. 1, shows several peaks that can be assigned to absorption bands of Dy^{3+} and Eu^{3+}

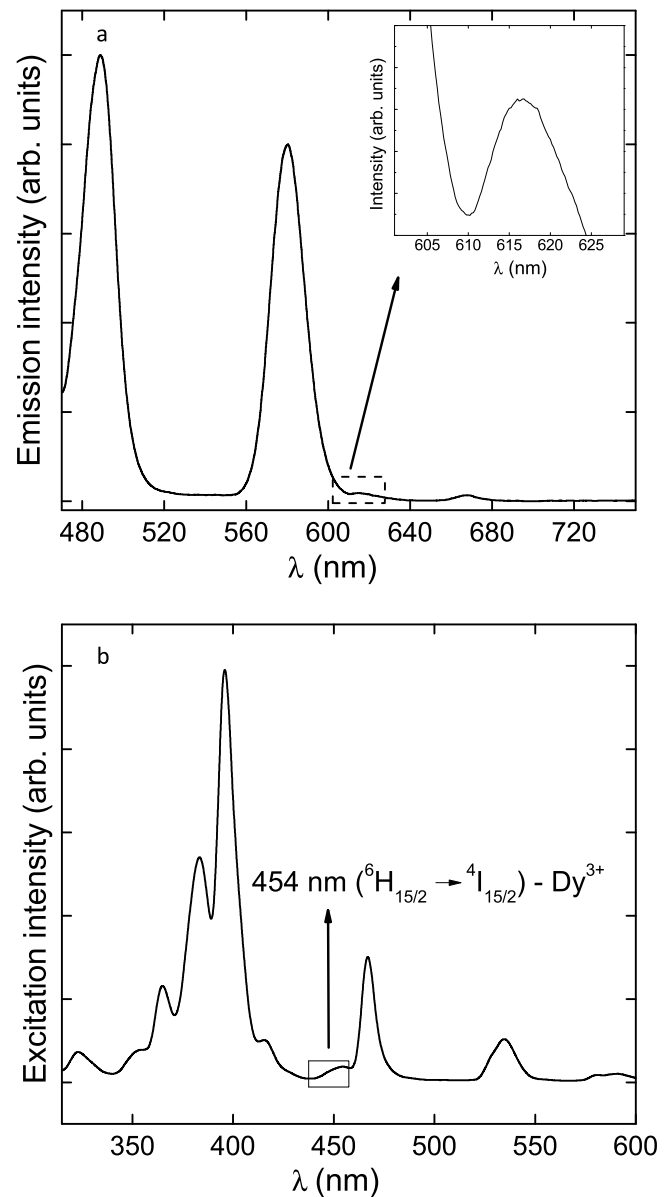


Fig. 2. (a) Emission spectra of the BBS-DyEu glass sample excited at 442 nm. The inset shows a zoom in of the lowest emission band centered at 616 nm, which is attributed to the ${}^5\text{D}_0 \rightarrow {}^7\text{F}_2$ transition of Eu^{3+} . (b) Excitation spectrum of the sample monitored at 616 nm.

ions. As labeled in Fig. 1, most of the bands are from the Dy^{3+} ion, which presents absorption at 349 nm (${}^6\text{H}_{15/2} \rightarrow {}^6\text{P}_{7/2} / {}^4\text{M}_{15/2}$), 364 nm (${}^6\text{H}_{15/2} \rightarrow {}^4\text{I}_{11/2}$), 386 nm (${}^6\text{H}_{15/2} \rightarrow {}^4\text{F}_{7/2} / {}^4\text{I}_{13/2} / {}^4\text{M}_{19/2,21/2} / {}^4\text{K}_{17/2}$), 424 nm (${}^6\text{H}_{15/2} \rightarrow {}^4\text{G}_{11/2}$), 453 nm (${}^6\text{H}_{15/2} \rightarrow {}^4\text{I}_{15/2}$), 747 nm (${}^6\text{H}_{15/2} \rightarrow {}^6\text{F}_{1/2} / {}^6\text{F}_{3/2}$), 797 nm (${}^6\text{H}_{15/2} \rightarrow {}^6\text{F}_{5/2}$) and 890 nm (${}^6\text{H}_{15/2} \rightarrow {}^6\text{H}_{5/2} / {}^6\text{F}_{7/2}$), while Eu^{3+} is responsible for the absorption band at 393 nm (${}^7\text{F}_0 \rightarrow {}^5\text{L}_7$) [27,28].

Fig. 2(a) displays the emission spectrum of the BBS-DyEu glass sample, excited at 442 nm. The two major emission bands, locate at 488 nm and 579 nm are attributed to the transitions ${}^4\text{F}_{9/2} \rightarrow {}^6\text{H}_{15/2}$ (characteristic blue emission) and ${}^4\text{F}_{9/2} \rightarrow {}^6\text{H}_{13/2}$ (characteristic yellow emission) of the Dy^{3+} ions, respectively. The inset in Fig. 2(a) highlights the lowest emission band centered at 616 nm, which is attributed to the ${}^5\text{D}_0 \rightarrow {}^7\text{F}_2$ transition of Eu^{3+} (characteristic red emission) [28,29]. Another weak transition at 667 nm (characteristic red emission) can also be observed in the emission spectrum, being assigned to the ${}^4\text{F}_{9/2}$

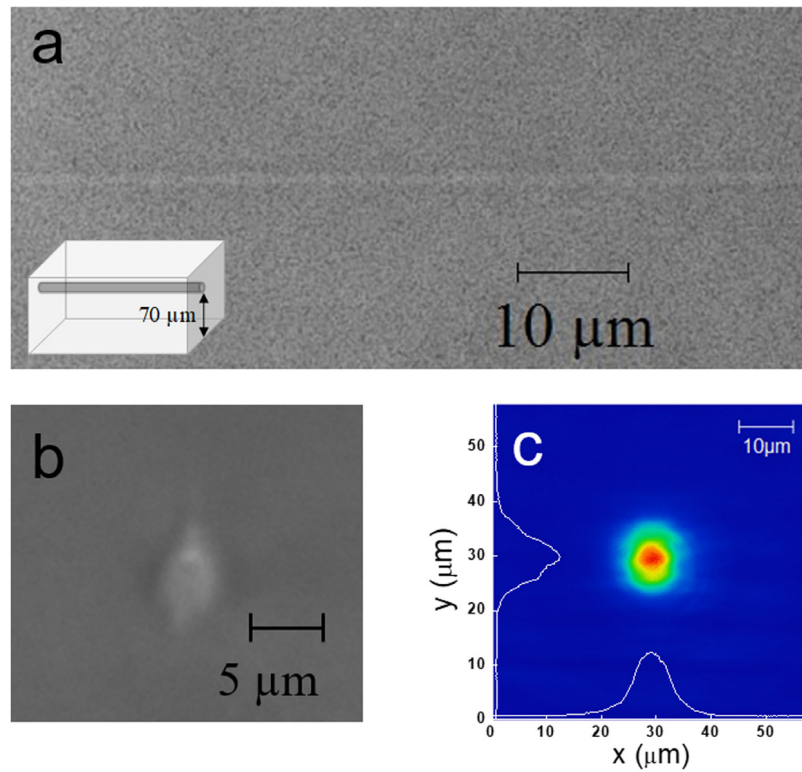


Fig. 3. Optical microscopy images of waveguides produced in the BBS-DyEu sample: (a) top view image of the fabricate waveguide. The inset illustrates the layout of the waveguide (b) cross-section view. (c) Near-field output profile of the light guided at 632.8 nm with its horizontal and vertical profiles.

→ ${}^6\text{H}_{11/2}$ transition of Dy^{3+} [30]. The same behavior was observed for excitation at 325 nm.

Glassy systems co-doped with Dy^{3+} and Eu^{3+} ions are known to support energy transfer process of non-radiative nature [31,32], which can be inferred through the excitation spectrum shown in Fig. 2(b), while monitoring the Eu^{3+} emission at 616 nm. Besides the expected excitation bands of Eu^{3+} , it is possible to observe a shoulder at 454 nm (marked with a rectangle) which is related to the characteristic excitation of the Dy^{3+} ion [33]. Such result indicates the energy transfer from Dy^{3+} to Eu^{3+} and it is likely to occur given the partial overlap between the emission bands at 488 nm (${}^4\text{F}_{9/2} \rightarrow {}^6\text{H}_{15/2}$) and 579 nm (${}^4\text{F}_{9/2} \rightarrow {}^6\text{H}_{13/2}$) of Dy^{3+} ions with the absorption bands of Eu^{3+} at 467 nm (${}^7\text{F}_0 \rightarrow {}^5\text{D}_2$) and 580 nm (${}^7\text{F}_0 \rightarrow {}^5\text{D}_0$), not assigned in the absorption spectrum (Fig. 1) due to its low intensity [28,29].

Fig. 3 shows optical microscopy images of the top (a) and cross-section (b) views of a waveguide produced by fs-laser micromachining in the BBS-DyEu sample. The 7.8-mm long waveguides were produced at approximately 70 μm below the sample surface (as illustrated in the inset of Fig. 3(a)), using pulse energy of 32 nJ and scanning speed of 10 $\mu\text{m}/\text{s}$. Such images reveal that the fabricated waveguides are homogeneous along their length and present a slightly elliptical cross-section, sizing approximately 5 μm .

A typical mode profile of light guided at 632.8 nm is shown in Fig. 3(c), demonstrating that the produced waveguide supports single-mode guiding with a nearly Gaussian intensity profile. By fitting such intensity distribution, assuming the fundamental mode of a step-index waveguide, a refractive index change on the order of 9.0×10^{-4} was determined, in agreement with other results reported in the literature [34–37].

Transmission measurements performed in the waveguides (7.8 mm long) shows that the overall loss, including coupling and propagation losses, is (7 ± 1) dB at 632.8 nm. By calculating the mismatch coefficients using the method described in Refs. [38,39], a propagation loss of (3 ± 1) dB/cm can be determined, which is in agreement with other ones reported by fs-laser written waveguides [40,41]. A fine tuning

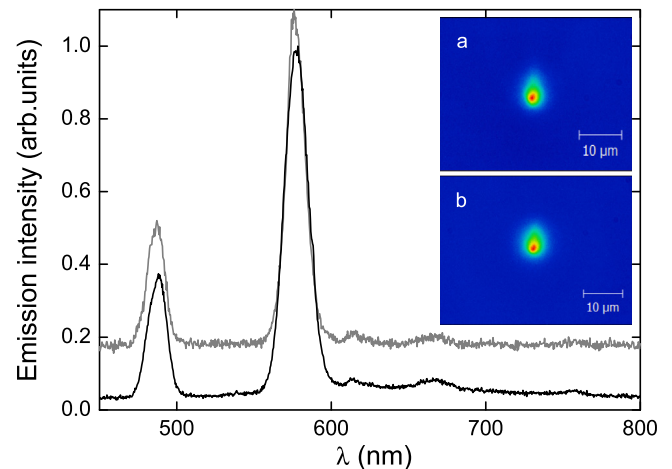


Fig. 4. Emission spectra obtained at the waveguide output when coupling light at 442 nm (black line) and 325 nm (gray line). The near-field output profile of the light guided at 325 nm (a) and 442 nm (b).

of the fabrication parameters, such as pulse energy, repetition rate and focalization can lead to an improvement in the waveguide propagation loss.

Using the objective lens coupling system previously described [26], light from a HeCd laser was coupled into the waveguide. Fig. 4 displays the spectra of the light collected at the waveguide output, for excitation at 442 nm (black) and 325 nm (gray). The peaks observed correspond to the emission of Dy^{3+} and Eu^{3+} ions, in agreement with the spectrum presented in Fig. 2(a). The insets in Fig. 4 show the near-field output profile of the guided sample emission when light at 325 nm (a) and 442 nm (b) were coupled. The observed intensity profile, in both cases, exhibits a nearly Gaussian distribution. While the 325 nm light is

completely absorbed at the beginning of the sample (~1.9 mm), for 442 nm the interaction length is on the same order of the waveguide length. For both cases, filters were used to block the excitation wavelength. Therefore, the observed spectrum and profile correspond only to the guided emission.

4. Conclusion

This study shows that femtosecond laser micromachining enables the fabrication of homogeneous waveguides in barium borate glass co-doped with rare earth ions (Dy^{3+} and Eu^{3+}), which are relevant for white light generation. The results demonstrate that the waveguides are able to support single-mode guiding of the sample emission when UV light is coupled. The waveguide characterization, performed with a He–Ne laser at 632.8 nm, revealed a refractive index change on the order of 9.0×10^{-4} , with an approximately Gaussian intensity profile and propagation loss of (3 ± 1) dB/cm. Finally, those waveguides are able to guide the emission of the Dy^{3+} and Eu^{3+} ions, resulting in an optical device with broad visible emission.

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