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405 nm pumped Ce³⁺-doped silica fiber for broadband fluorescence from cyan to red

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ABSTRACT

A pure Ce-doped silica fiber is fabricated using modified chemical vapor deposition (MCVD) technique. Fluorescence characteristrics of a Ce-doped silica fiber are experimentally investigaed with continous wave pumping from 440 nm to 405 nm. Best pump absorption and broad flourescence spectrum is observed for ~ 405 nm laser. Next, the detailed analysis of spectral response as a function of pump power and fiber length is performed. It is observed that a -10dB spectral width of ~ 280 mm can be easily achieved with different combinations of the fiber length and pump power. Lastly, we present, for the first time to the best of our knowledge, a broadband fluorescence spectrum with -10dB spectral width of 301 nm, spanning from ~ 517.36 nm to ~ 818 nm, from such fibers with non-UV pump lasers.

Keywords: Ce-doped fiber, broadband spectrum, rare earth doped, Ce-ion

1. INTRODUCTION

 Ce^{3+} -doped and Ce^{3+} - Ln^{3+} -codoped crystals, silicate, phosphate and fibers are of great interest for their broadband fluorescent emission across the visible and near IR wavelength range . Ce^{3+} ions have been investigated in aforementioned materials as independent luminescence centers and sensitizers. These Ce-doped materials has been used as scintillators and light emitters exploiting the allowed 5d-4f transition in Ce^{3+} ions.^{1,2} On one hand radiation detection has been the motivation to forward the research on scintillators;^{3,4} and on the other hand Ce-doped silica fiber has been studied for non-invasive biomedical technique such as high resolution optical coherence tomography (OCT).^{5,6} At the same time, Ce has been used with phosphors to achieve white light from light-emitting diodes.⁷ However, due to excellent fluorescence spectrum promise of Ce-doped silica fibers they may be employed in future for both visible light emission and possibly supplement greatly exciting and rapidly growing field of visible light communication. Towards this the primary requirements for such fibers would be to ideally use non-UV pump sources and provide broadband emission spectrum across the visible part of the optical spectrum.

In this study, fluorescence properties of a Ce-doped silica fiber are experimentally investigated. The doped fiber is prepared using modified chemical vapor deposition (MCVD) technique. The fluorescence spectra of the doped fiber is examined under non-UV 405 nm continuous wave pump laser source. Fluorescence spectrum is observed to be strongly affected by the length of the fiber both in shape and peak emission wavelength. We also demonstrate, possibly for the first time, a broadband spectrum with a -10dB spectral width of 301 nm from such fibers.

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Figure 1. Cross-section of Ce-doped fiber.



Figure 2. The proposed energy transfer mechanism for Ce^{2+} in Ce-doped fiber.

2. EXPERIMENTAL DETAILS

2.1 Fabricating Cerium-doped fibers

The Ce-doped silica cladding fiber is drawn from the preform fabricated by standard MCVD technology. The technological process of the preform fabrication consists of several principle steps. At the first step, the inner part of high purity silica substrate tube is deposited by powder like "soot" layer including SiCl₄ and fluoride. This process is accompanied by simultaneous burning at the high temperature in the range of 1500-1700°C. Introduction of fluoride ions allows to decrease the refractive index and form internal waveguide structure. At the next step, the rare-earth solution composed of 1 mol % of Ce(NO₃)₃ · 12H₂O is introduced into the tube. The following sintering of the tube to evaporate the liquid from rare-earth dopant solution is carried at a temperature of 500° C. Finally, the tube is collapsed and sealed.⁸ Figure 1 shows the cross-section of Ce-doped fiber. The fiber has near ideal circular shape with outer cladding and core diameters of $\approx 146 \ \mu$ m and $\approx 13 \ \mu$ m, respectively.

2.2 Characterization

We suggest that the active fiber contains trivalent and divalent Ce ions. Upon absorbing blue light an electron in the trivalent Ce ions is excited from lower levels manifold of Ce ion $({}^{2}F_{5/2} \text{ and } {}^{2}F_{7/2})$ to upper levels $({}^{2}D_{3/2} \text{ and } {}^{2}D_{5/2})$. The electron can transit to the ground state of Ce ion emitting light in blue and green wavelength range. Due to the absence of allowed energy states between 4f-5d orbital, the emitting properties of trivalent Ce



Figure 4. Fluorescence Spectrum at different pump wavelengths.

ion is limited in short part of the visible spectrum range. Therefore, Ce^{3+} ions mostly takes part in pump light absorption and emission/re-absorption of wavelength below ≈ 550 nm, whereas, the luminescence properties of the fiber investigated in this article can be explained by down-conversion mechanism primarily occurred in Ce^{2+} sites. The Figure 2 shows the energy level diagram which is currently proposed to explain the intra-configuration transition of Ce^{2+} ions in the fiber.⁹Divalent Ce characterizes by rich energy level structure with energy gap from 10 000 till 25000 cm⁻¹, which allows to cover all visible and short wavelength part of near-IR wavelength range. However, to efficiently exploit Ce^{2+} energy level structure it is necessary to minimize the parasitic non-radiative relaxation rate, which is high in pure silica glass host due to large phonon energy value ≈ 1100 cm⁻¹. Therefore, the energy gap between upper excited and lower excited energy levels must be at least 4 phonons wide. As a consequence, any potential emitting levels in silica glass should be 4400 cm⁻¹ above lower lying levels.¹⁰

To explore the quenching effect by multiphoton non-radiative relaxation and define the optimum energy transfer condition from the ground state to the upper excited states of Ce^{2+} ions, we experimentally investigate several pump sources with the optical excitation wavelengths of 440 nm, 420 nm, 416 nm, 414 nm, 413 nm, 405 nm. The excitation lasers used are TO-Can packaged diode lasers with continuous wave (CW) output powers of > 5 mW. The excitation lasers were temperature controlled at 20 degC and their CW output is coupled to the fiber with a 1:1 A-coated aspheric lens assembly. The other end of the Ce^{3+} -doped fiber is butt coupled to a multimode fiber, "QP-1-VIS-NIR", with core diameter of 600 μ m. A schematic for the experimental setup is presented in Figure 3. The output of the multimode fiber is analyzed using a "Labsphere CDS-600" spectrometer and "SpectralSuite" software. The output optical power and spectrum were measured alternatively in that order under the same excitation conditions.



Figure 5. Fluorescence output power vs fiber length and pump power at 405 nm.



Figure 6. Measured spectra at different pump powers, obtained at fiber lengths of 132.6 cm (30.6 mW), and 142.7 cm (94.8 mW)

3. RESULTS AND DISCUSSION

To characterize fluorescence spectrum of ce-doped fibers it is first measured at different pump wavelengths, in the blue region of the optical spectrum, starting with 440 nm and moving towards shorter wavelengths. The fiber length used to identify the optimum pump wavelength is ~ 70 cm and the pump power is kept between 12 mW to 18 mW. Figure 4 depicts the fluorescence spectrum at selective pump wavelengths (refer to section 2.2). Following the trend on increased fluorescence with shorter pump wavelength it was observed that with \sim 14 mW of 405 nm pump we observe the most efficient pump-to-signal wavelength conversion at 70 cm length of the fiber.

Next, the behavior of fluorescence spectra as a function of fiber length and pump power is investigated to determine the optimal parameters for broadband fluorescence. Initially spectral measurements for a 170 cm long fiber is performed at different discrete pump powers of 30 mW, 62.5 mW, 94.8 mW and 127.4 mW. Spectral measurements at the same pump powers is then performed for different fiber lengths by slicing the fibers in the steps of ≈ 10 cm with the shortest fiber length of 30 cm. The fiber is sliced from the end used for butt coupling as this allowed for consistency in recording output optical power at the far end of the ce-doped fiber due to large core diameter of the multimode fiber.

The measured fluorescence output power (excluding the residual pump at shorter fiber lengths) as a function of fiber length at different pump powers used is shown in Figure 5. The output power gradually increase with decrease of the fiber length reaching >300 nW for ≈ 30 cm of fiber length pumped with >120 mW pump power.

Figure 6 depicts the observed fluorescence spectrum for two different fiber lengths at different pump powers. A correlation is observed between pump absorption and fiber length which is almost independent of applied pump power. The pump absorption is estimated from the measurement data to be $\sim 2 \text{ dB/cm}$. For fiber lengths > 58 cm all pump is absorbed by the fiber, irrespective of optical pump power. It is also observed that fluorescence



Figure 7. (a) Fluorescence bandwidth vs fiber length (>100 cm) and pump power: (a) peak wavelength (b) full-width at -10 dB.

spectrum shifts towards longer wavelengths with increasing fiber length and the spectral shape is also affected. The observed shift in peak emission wavelength is >80 nm as a function of fiber length irrespective of pump power, see Figure 7a.

Furthermore, the fluorescence spectra due to 5d-4f transitions on Ce-ions is broad as one would expect. We have observed that the spectral width over the whole range of fiber length and pump power used, increases almost linearly with increasing fiber length until a maximum is reached. The behavior has no observable correlation with pump power used. This -10 dB spectral width is >230 nm. A -10 dB-bandwidth of ~ 280 nm (see Figure 7b) is observed for different fiber lengths. A -10 dB-bandwidth of >295 nm (see Figure 7b) is observed for fiber lengths of ~132 cm To the best of our knowledge, this is the widest spectral bandwidth demonstrated from such fibers.

In conclusion, we demonstrate broadband, visible to near-IR, fluorescence from the pure Ce-doped silica fiber with -10dB spectral width of >295 nm with peak wavelength ~ 643 nm and peak power density of 0.06 nW/nm.

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