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Spectroscopy of Yb:Tm doped tellurite glasses for efficient infrared fiber laser

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ABSTRACT

We performed extensive spectroscopy of tellurite glasses doped with high concentration of Tm ions for laser emission at around 2 micron wavelength. The aim of the work is to develop a glass suitable for single-frequency fiber laser. In fact such a kind of laser require the use of short cavity length and therefore high gain per unit length medium. Tellurite glasses allows high-doping concentration and are therefore an excellent candidate. In these paper we review our recent results. In particular we address the optical and thermo-mechanical properties of several tellurite glasses (75mol%TeO₂.20mol%ZnO. 5mol%Na₂O) with Tm³⁺ doping up to 111,564 ppm.

Keywords: tellurite glasses, Tm-doping, fiber laser

1. INTRODUCTION

Laser sources operating in the 2 μm wavelength region are nowadays of great interest due to potential applications ranging from laser surgery based on water absorption of tissue, to remote sensing of some atmospheric molecular species and Lidar applications [1]. Among applications the two latter types as well as many others will benefit from relatively high-power single frequency sources. The output power and narrow linewidth will improve the overall detection sensitivity and system performance.

An extensive investigation have been carried out in the last decade on fiber based laser devices using Tm³⁺ doped glass and operating in the 2 μm region region [2]-[8]. Various glass hosts have been investigated and laser operation have been demonstrated in several glasses comprising silicate, germanate, phosphate and tellurite glasses.

However the majority of these works however have concerned low concentration levels of Tm³⁺ ions in glass, typically of the order of 1 wt%. Up to know, only highly doped Tm³⁺ germanate glasses (up to 10 wt%) have been reported together with the lasing demonstration from a 4wt% Tm doped germanate glass fiber[5,6].

We believe tellurite glasses could offer a better alternative to germanate glasses for the specific application we target. Their lower phonon energy and high rare-earth ion solubility are direct advantages in order to meet the required specification.

We review here our recent results investigation of highly doped Tm³⁺ tellurite glass to develop short cavity fiber laser devices operating in the 2 μm region. We developed a series of tellurite glasses with composition containing up to ~110 000 ppm in Tm³⁺ ions. We analyzed the optical and thermo-mechanical properties and the results are reported and discussed.

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2. GLASS FABRICATION

The composition of the tellurite host glass was the following: 75 mol% TeO₂.20mol% ZnO.5mol%Na₂O (indicated as T0 in Table 1) [9]. All glasses were prepared by melt quenching from mix powder batches, inside a glove box in a dry atmosphere with water content of about 7 ppm. The chemicals employed (together with their purity) were the following: TeO₂ (99.99%), ZnO (99+%), Na₂CO₃ (99.995%), Tm₂O₃ (99.99%). Relative molar ratio of the host glass constituent oxides was kept the same for all samples, regardless of Tm doping. The fabricated samples were based on the following notation: (100- x) (75TeO₂ - 20ZnO - 5Na₂O), where x = 0.36, 0.72, 1.08, 2.14, 3, 4, 5, 6, 7, and 10 %mol of Tm³⁺. Glass melting was carried out in Pt crucibles at around 900 °C for 2–3 h, then pouring on a preheated brass plate at 300 °C and annealing followed. The whole process required around 20 h of operation [9].

Glass label	Tm ³⁺ concentration		T _g ±3°C	T _x ±3°C	ΔT= T _x -T _g ±6°C	n ±0.001 at 1533 nm
	ppm	(×10 ²⁰ ions/cm ³)				
T0	0	0	303	417	117	1.996
T0.72	8690	1.65	310	442	132	1.994
T1.08	12,993	2.47	313	447	134	1.993
T2.14	25,508	4.87	317	463	146	1.988
T3	35,494	6.84	322	468	146	1.987
T4	46,920	9.06	321	473	152	1.983
T5	58,151	11.3	320	469	149	1.979
T6	69,194	13.5	326	473	147	1.976
T7	80,052	15,7	330	479	149	1.973
T10	111,564	22.1	333	466	133	1.962

Table 1. List of glasses manufactured. T0 is the glass base.

3. GLASS CHARACTERISATION

We measured the characteristic temperatures of all glasses manufactured. In table 1 we report the T_g (glass transition temperature), T_x (onset crystallization temperature), the glass stability parameter ΔT= T_x-T_g and the refractive index at 1533 nm, the longer test wavelength available. The glass transition temperature and the onset crystallization temperature were measured using a commercial Differential Scanning Calorimeter (DSC) from Perkin Elmer with a measurement error of ± 3 °C. From Table 1 we can note an increase of the glass transition temperature with respect to the increase of the Tm³⁺ content. The glass stability parameter ΔT= T_x-T_g shows the stability depends upon the host composition. A minimum glass stability value of 132 °C was obtained for a concentration in Tm³⁺ of 8690 ppm for sample T0.72 while the most stable glass compositions T4 shows a stability parameter of 152 °C. This difference can be explained by the Raman spectra measurements, shown in Fig1., suggesting a change in glass structure due by the incorporation of Tm ions [9]

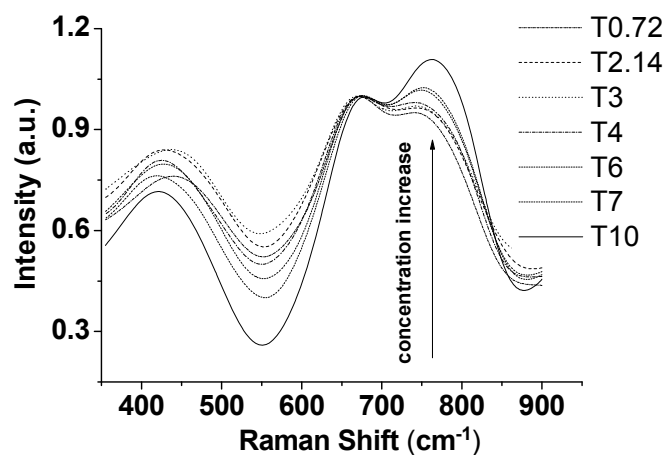


Figure 1. Raman spectra normalized to the peak at 650 cm^{-1} . Excitation wavelength was 1064 nm [From Ref.9].

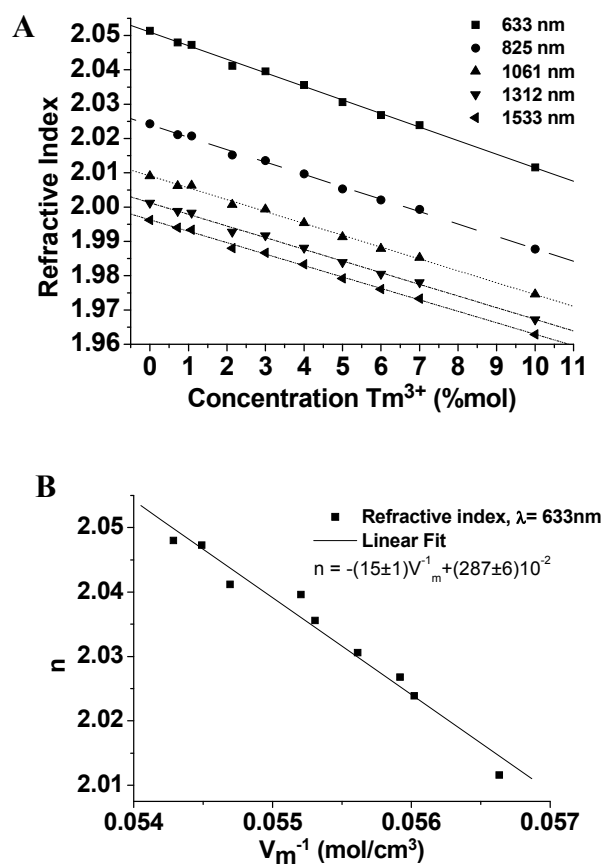


Figure 2. A) Refractive index as a function of Tm concentration; B) Refractive index versus the molar volume.

To characterize the glass refractive index, a main parameters for fiber design, we performed a deep investigation as shown in Fig. 2A where refractive index (n) values at five different wavelengths (633 nm, 825 nm, 1061 nm, 1312 nm, 1533 nm) for different Tm^{3+} concentrations are reported. Data were obtained by prism coupling technique (Metricon, model 2010). The resolution of the instrument was of ± 0.0001 . In addition the influence of molar volume (V_m) on the refractive index (n) can be seen in Fig. 2B, where linear correlation is proved by least square fit.

4. GLASS SPECTROSCOPY

The absorption cross section obtained for T10 by FTIR spectroscopy is shown in Figure 3. In order to identify an optimum glass composition for a 2 μm laser application, a systematic investigation of the 1.8 micron fluorescence from the 3F_4 level of excited Tm ions was carried out. We used polished 1 mm thick samples. All samples were excited using 200mW at 781 nm from tunable laser source (mod. SDL 8630). The Fluorescence spectra were recorded using a OceanOptics NIR256 spectrometer. In all measure we carefully tried to avoid re-absorption of the emission from excited Tm^{3+} ions by surrounding Tm^{3+} ions. The pump beam was free space with $M^2=1.1$ and was carefully focused on the sample edge.

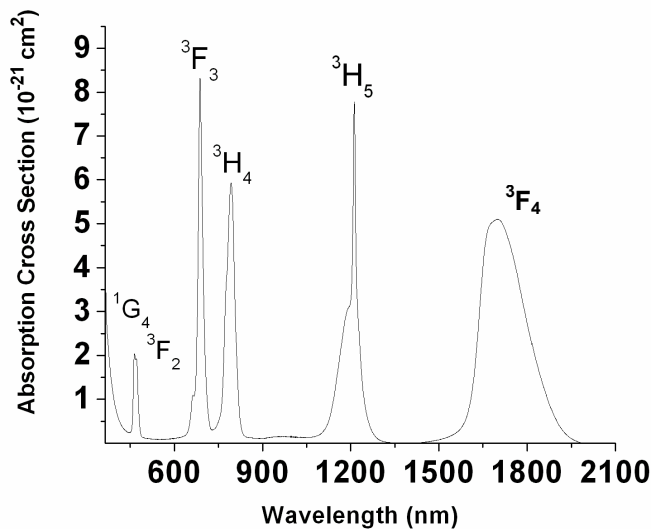


Figure 3. Absorption spectra of glass T10.

In Fig.4 is depicted the Tm energy level system and the two main fluorescence emission lines at 1.82 μm and at 1.46 μm .

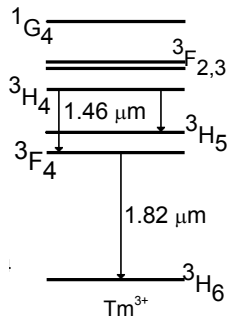


Figure 4. Tm energy levels system and main fluorescence emission lines

Figure 5 shows the emission spectra of the Tm^{3+} doped glass presented in Table 1. The two emissions peaks at around $1.463 \mu m$ and $1.822 \mu m$ appear clearly. The peak at around 1580 nm that appears for low doped samples is a spurious one and it originates from the pump sources.

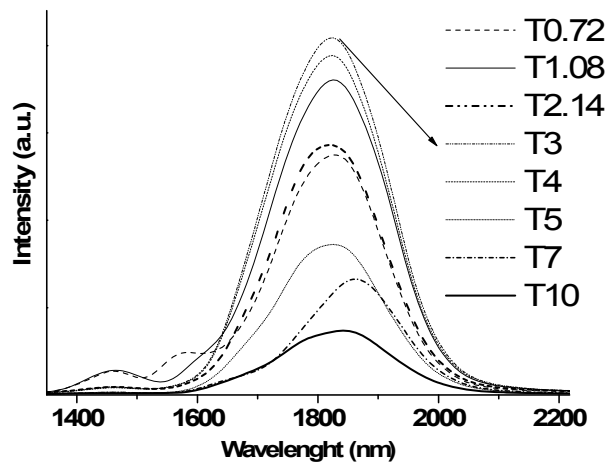


Figure 5. Emission spectra [9] of glasses listed in Table 1.

We note a decrease in the intensity of the $1.4 \mu m$ emission with increasing Tm concentration while the intensity of the $1.8 \mu m$ emission initially increases up to the T3 sample and then decrease as Tm^{3+} concentration increasing. We are investigating the trend in order to extract the Tm main parameters such as energy-transfer among excited ions and cross-relaxation processes as a function of the doping levels.

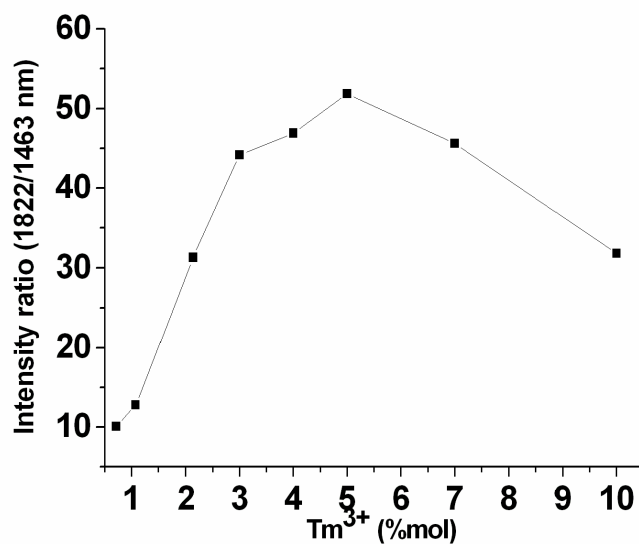


Figure 6. Emission ratio (${}^3\text{H}_4 \rightarrow {}^3\text{F}_4$)/ (${}^3\text{F}_4 \rightarrow {}^3\text{H}_6$) of the two main fluorescence peaks.

Figure 6 shows the ratio between the peak intensity of the 1.8 μm emission and the 1.46 μm emission versus Tm concentration. This ratio reaches a maximum for a concentration of 58 151 ppm (sample T5). We can consider this ratio as a rough figure of merit indicating that Tm ions mainly store the inversion in the upper laser level. With this respect concentration around 60000 ppm should be very promising. We are however still investigating the optimization issues. Further comments on doping concentration optimization are reported in the next section.

5. LIFETIME MEASUREMENTS

We performed a set of measurement of the lifetime of the ${}^3\text{F}_4$ level by measuring the decay of the 1.8 μm fluorescence. Figure 7 shows as example the decay curves for three samples: T1.08, T3 and T10 [9]. In Figure 8 are reported the measured values for the glasses listed in Table 1 [9]. The measurement were done by on/off modulation of the 780 nm pump diode with 10 Hz repetition rate and 50% duty cycle. The on and off intervals of 50 ms were long enough to reach steady-state inversion and complete depletion, respectively.

Fitting of the curve of lifetime values is also reported in Fig.8 vs. Tm concentration. The fitting gives a Tm³⁺ quenching concentration value of around 2-3 mol% with a lifetime fit parameter of $\tau_0 = 2.8\text{ms}$. From Fig. 8 we can also see that apparently after T4 sample (4 mol%) we have a strong decrease in the measured lifetime. Considering it together with the measures reported in Fig.5 we believe that optimum Tm concentration would be in between 4 %mol and 5 %mol.

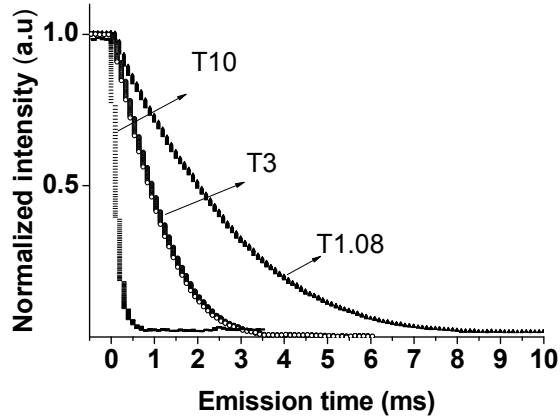


Figure 7. Decay curve of 1.8 micron fluorescence

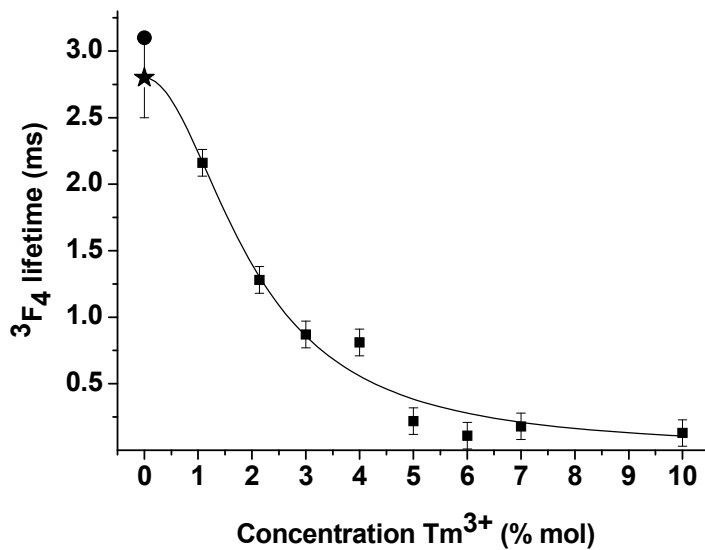


Figure 8: Lifetime of 3F_4 level.

6. CONCLUSION

This paper presents spectroscopic and optical properties of highly Tm^{3+} doped tellurite glasses. All prepared glass samples were homogeneous, bubble free and their thermal stability data were in the average of tellurite glasses. We measured a lifetime quenching Tm^{3+} concentrations higher than 2 mol% with maximum predicted lifetime value of around 2.8 ms. By evaluating the fluorescence spectra and the lifetimes values we believe the optimum Tm doping

concentration will range from 4 mol% to a maximum of 5 mol%. This doping level is suitable to build compact single-frequency lasers emitting at around 2 micron wavelength.

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