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**Tm<sup>3+</sup> and Yb<sup>3+</sup> co-doped tellurite glasses for short cavity optical fiber lasers: fabrication and optical characterization**

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**Keywords:** tellurite glass, Tm, Yb, rare earth.

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**Abstract:**

In this paper Tm-doped tellurite glasses (75TeO<sub>2</sub>- 20ZnO-5 Na<sub>2</sub>O, mol%) were studied and sensitization with Yb<sup>3+</sup> as co-dopant was carried out in order to assess its influence on the emission properties and improve pump efficiency and wavelength emission range. Emission spectra and lifetime measurements were obtained by pumping either at 785 nm or 980 nm, thus exploiting the Yb-Tm energy transfer mechanism. Doping levels of Yb from below 1 to up 5 mol% were investigated in order to evaluate the effectiveness of energy-transfer process and glasses containing an overall content of 9 mol% rare earth ions were proved to be successfully fabricated. All glasses were fully characterized from a material and spectroscopic point of view. Tellurite glasses with 4 and 1 mol% of Tm<sup>3+</sup> and Yb<sup>3+</sup> ions respectively were found to show a good compromise between emission efficiency and lifetime decay values. A quenching concentration of Yb<sup>3+</sup> ions up to 14 mol% was also defined for this type of glass system.

**1. Introduction**

The recent interest in lasers emitting in the near infrared wavelength range has stimulated the investigation of novel materials able to easily incorporate high amounts of rare earth ions, thus making possible the fabrication of short cavity compact and single frequency coherent sources. In particular the emission wavelength range between 1.9 and 2 micron is of great interest for various applications, including remote sensing and LIDAR systems [1].

Tm<sup>3+</sup> is a suitable ion for emission at around 2 microns and recent research activities dealt with Tm-doped silica, fluoride and tellurite glasses and optical fibers [2;3;4;5;6;7]. Among possible glass hosts, tellurite glasses combine the lowest phonon energy among oxide glasses with the higher chemical stability towards non oxide glasses [8;9]. Besides, their ability to incorporate high amounts of rare earth ions make them suitable candidates for the development of short compact fiber laser sources for sensing applications. Yb can be used as a sensitizer and it allows pumping with more powerful and less expensive laser diodes operating at 980 nm. Previous works investigated Yb/Tm co-doped tellurite glasses [10;11; 12], but doping levels were limited to a maximum of few mol %. Tm and Yb/Tm doped fiber lasers have been recently demonstrated, however no investigation dealt with high

concentrations of Tm and Yb.

Aim of this paper is to investigate the effect of Yb<sup>3+</sup> co-doping on Tm<sup>3+</sup> ions spectroscopic properties when their content is higher than 2 mol% and thus allow the identification of a good candidate active material for short cavity fiber lasers.

## 2. Experimental procedure

### 2.1. Glass fabrication

Glasses were prepared by melt quenching from mix powder batches, inside a glove box in a dry atmosphere with water content of about 10 ppm. The chemicals employed (together with their purity) were the following: TeO<sub>2</sub> (99+%), ZnO (99.99+%), Na<sub>2</sub>CO<sub>3</sub> (99.995%), Tm<sub>2</sub>O<sub>3</sub> (99.99). The fabricated glasses were based on the following host mol% composition: 75TeO<sub>2</sub>:20ZnO:5Na<sub>2</sub>O. A series of Yb and Yb/Tm doped samples were then fabricated while keeping the relative molar ratio of the host glass constituent oxides constant. The glass samples were divided into three groups: group I glasses were Yb-doped glasses doped with 0.5, 1, 3 and 5 mol% of Yb<sup>3+</sup> with respect to oxide glass constituents; group II glasses had a constant 1 mol% Yb<sup>3+</sup> content, whilst Tm<sup>3+</sup> varied from 1 to 2 to 4; group III was characterized by keeping a constant amount of Tm<sup>3+</sup> ions (4 mol%) and varying the Yb<sup>3+</sup> amount from 1 to 3 to 5 mol%, respectively. Acronyms of the glass samples follow the dopant initial letter together with the number of the mol%, as reported in Table 1.

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 at around glass T<sub>g</sub> followed.

### 2.2 Glass characterization: thermal and physical properties

Thermal analysis was performed on fabricated glasses using a Perkin Elmer DSC-7 differential scanning calorimeter up to 550°C under Ar flow with a heat rate of 10°C/min in sealed Al pans using 30 mg glass samples. Thermal analysis was employed to determine the effect of glass composition on glass stability which can be measured with the quantity T<sub>x</sub>-T<sub>g</sub> (T<sub>x</sub> is crystallization peak onset values and T<sub>g</sub> is glass transition temperature). A random error of ±3°C was observed.

Density measurements were obtained by precise mass measurements in air and water environment on

selected specimens following the Archimede's principle.

### *2.3 Glass characterization: optical properties*

Glasses were cut into 1 mm thick slices and polished to an optical quality. UV-VIS transmission spectroscopy was carried out in order to assess the absorption spectra of the rare earth doped glasses. Refractive index was measured for all samples at three different wavelengths (825, 1061 and 1533 nm) by prism coupling technique (Metricon, model 2010). The resolution of the instrument was of  $\pm 0.0001$ . Ten scans were used for each measurement. Standard deviation in refractive index at different point of the same sample was around  $\pm 0.0003$ .

Lifetime measurements of Tm  $^3F_4$  excited level were carried out by pumping either at 785 nm (LMA Raman diode Laser, mod. L#35478, excitation power of 100 mW) or at 978 nm (SDL 81236 diode laser, excitation power of 350 mW). Fig. 1 shown the energy levels of Yb $^{3+}$  and Tm $^{3+}$  ions. For lifetime decay analysis, excitation power was on-off modulated with repetition frequency of 20 Hz using either a chopper (in the case of the 785 nm wavelength source) or a pulse signal generator. All signals detected were filtered to remove the residual pump and a SPEX monochromator was used to select the desired wavelength. A Hamamatsu PbS detector was employed and the fluorescence spectra and lifetime decay signals were collected 90° away from the incident to avoid re-absorption. Decay curves were fitted exponentially. Experimental and fitting errors were taken as  $\pm 0.02$  ms.

## **3. Results**

### *3.1 Glass characterization: thermal and physical properties*

All prepared glasses were homogeneous and X-Ray analysis on powdered samples showed no occurrence of diffraction peaks assignable to crystalline phases. Thus all samples were considered suitable as candidate glasses for optical fiber cores. The rare earth content and concentrations of the prepared glasses are shown in Tab. 1, together with their density, refractive index values and their main thermal properties, measured using DSC analysis ( $T_g$ ,  $T_x$ ). Density and  $T_g$  generally increased with increasing rare earth content, as previously observed [7] on Tm-doped tellurite glasses.  $T_x$  values are also following a similar behavior, except for group III glasses, where a decrease in  $T_x$  was

observed. Group I and II glasses showed an increase in stability versus de-vefrification with increasing Yb and Tm content, respectively. Whilst group III glasses show a decrease in  $T_x-T_g$  values, but to a value not far from sample Y0.5.

### *3.2 Glass characterization: optical properties*

Refractive index values of Tm-doped prepared glasses (Group II and III glasses) are plotted in Fig. 2 and compared with data of our previous work on Tm-only doped tellurite glasses [7]. It is observed that the introduction of  $Yb^{3+}$  ions inside the glass results in a decrease of refractive index, in a similar way to the introduction of Tm. However Yb has a stronger effect than Tm on the decrease in refractive index: the slope of  $n$  versus  $Yb^{3+}$  mol% is -0.00417 for group I glasses and - 0.00478 for group III glasses (with a constant 4 mol% Tm content), whilst for  $Tm^{3+}$ -doped glasses the slope of  $n$  is -0.00334 for Tm-only doped glasses and - 0.00252 for group II glasses.

The UV VIS spectra of the prepared samples showed characteristic absorption transitions from the ground state, as displayed in Fig. 3 in the case of a group I (Y5) and a group III (T4Y3) glasses. The occurrence of the absorption peak of the transition  ${}^2F_{7/2} \rightarrow {}^2F_{5/2}$  in the case of the Tm/Yb doped glass allows pumping at around 980 nm. Fig. 4 shows absorption cross section in the near infrared region absorption related to  $Yb^{3+} {}^2F_{7/2} \rightarrow {}^2F_{5/2}$  transition.

### *3.3 Fluorescence optical spectra of prepared glasses*

Fluorescence spectra were collected for group II and group III glasses, which were excited at the wavelength of 980 nm, while samples containing  $Tm^{3+}$  ions as co-dopant were also excited at the wavelength of 785 nm. Fig. 5 and 6 show the fluorescence spectra of group II and III glasses by pumping at 980 nm. For group II glasses, there is a first increase in intensity of the  ${}^3F_4 \rightarrow {}^3H_6$  with doubling  $Tm^{3+}$  ion concentration from 1 to 2 mol%, followed by a decrease with sample T4Y1. A similar effect was observed for group III glasses, with increasing  $Yb^{3+}$  content.

Fig. 7 shows the intensity ratio between  ${}^3F_4 \rightarrow {}^3H_6$  and  ${}^3H_4 \rightarrow {}^3F_4$  emission transitions vs.  $Tm^{3+}$  concentration for  $TxY1$  glasses ( $x = 1, 2, 4$ ) by pumping at 980 nm. It can be seen that such value increases with increasing Tm content in the glass. The effect of  $Yb^{3+}$  ions content on the same

intensity ratio was also studied on group III glasses: Fig. 8 shows an opposite behavior if compared to group II, since it was observed to decrease linearly.

### *3.4 Lifetime decay analyses of prepared glasses*

Samples containing Yb<sup>3+</sup> ions (all glasses) were excited at the wavelength of 980 nm, while samples containing Tm<sup>3+</sup> ions as co-dopant (group II and III) were also excited at the wavelength of 785 nm. All lifetime values measured within this paper are reported in Tab.2. All decay curves were fitted exponentially.

#### *3.4.1 - 980 nm pumping*

Lifetime decays of Yb-doped glasses pumped at 980 nm are shown in Fig. 9. As evident, all curves show an exponential decay and such decay is observed to become stronger with increasing Yb<sup>3+</sup> ion content.

As example, the lifetime decay curves of group II glasses are reported in Fig. 10. It is clear that the profile is again exponential and with increasing Tm<sup>3+</sup> content, the Tm<sup>3+</sup>F<sub>4</sub> lifetime is seen to decrease. Tm<sup>3+</sup> ion decay curves were focused on the measurement of <sup>3</sup>H<sub>4</sub> and <sup>3</sup>F<sub>4</sub> lifetime values. Tm<sup>3+</sup>H<sub>4</sub> lifetime decay values proved to be difficult to collect, thus the determination of lifetime values was more critical. Tm<sup>3+</sup>F<sub>4</sub> lifetime measurements were easily collected thanks to a stronger signal, and thus clear decay curves were recorded.

#### *3.4.2 - 785 nm pumping*

Tm<sup>3+</sup>H<sub>4</sub> lifetime values were very difficult to collect, being the measurement taken at the very edge of the sample: the intensity of the signal was low and thus triggering with oscilloscope was not possible. In particular only T1Y1 glass provided a measurable value of lifetime. Tm<sup>3+</sup>F<sub>4</sub> decay curves gave strong signals and the decay curves of group II glasses are reported in Fig. 10 as an example.

#### 4. Discussion

All prepared glasses showed interesting physical properties and absence of XRD detectable crystalline phases, even for high amounts of rare earth ions: in our case we explored the highest amounts of rare earth concentrations in literature on tellurite glasses. Measured characteristic temperatures ( $T_g$ ,  $T_x$ ) are in line with Tm-doped tellurite glasses reported in literature and show a decrease in glass stability only when very high amounts of Tm and Yb ions are included in the glass systems.

Fluorescence spectra showed that Yb-codoping of Tm tellurite glasses does not affect the performance of the Amplified Spontaneous Emission (ASE): indeed the results shown in Fig. 7 are providing an evidence of the increase in emission intensity of the 1800 nm emission if compared to the 1470 nm emission. Whilst at high Tm concentration (namely at 4 mol%  $Tm^{3+}$ ), the increase in  $Yb^{3+}$  amount results in a decrease of the efficiency of the targeted 1800 nm emission. It is thus possible, on the basis of the measured fluorescence spectra, that T4Y1 would be the best glass in term of relative Tm and Yb contents.

Regarding lifetime decay analyses, a first interesting discussion is to be developed around the performance of  $Yb^{3+}$  ions. Indeed glasses doped only with  $Yb^{3+}$  ions (group I) show a continuous decrease in lifetime data, whilst in literature [11] an increase followed by a decrease in lifetime was observed, where the self-trapping process is evident [13]. In the case of the measurements carried out within this study, particular care has been taken in order to minimize re-absorption: in order to assess the quality of the measurements, a comparison with powdered samples was carried out and we succeeded in minimizing re-absorption.

In order to assess the effect of Yb doping concentration on its lifetime, the following equation was used [13]:

$$\tau = \frac{\tau_0}{1 + \frac{9}{2\pi} \left(\frac{N}{N_0}\right)^2} \quad (1)$$



where  $\tau$  is the lifetime of the rare earth ion,  $\tau_0$  is the single ion lifetime or radiative lifetime,  $N$  is the concentration of the rare earth ion in mol % and  $N_0$  is the quenching concentration of the rare earth ion within the host. Applying this formula to the lifetime data of group I glasses, by using as radiative lifetime a value of 0.46 ms the fitting appears as reported in Fig.11 for a quenching concentration  $N_0 = 14 \pm 1$  mol %. As evident the experimental value of Y0.5 glass appears to be slightly out of the fitting curve because its measured value is 0.5 ms. However the first data obtained through this fitting show great opportunities for the fabrication of highly doped Tm-tellurite glasses, also by using  $\text{Yb}^{3+}$  as sensitizer. Indeed high amounts of Yb do not significantly affect the  $\text{Yb } ^2\text{F}_{5/2}$  lifetime, providing an evidence of its efficient role as sensitizer for  $\text{Tm}^{3+}$ . However more work has to be carried to experimentally demonstrate the efficient incorporation of high amounts of  $\text{Yb}^{3+}$  ions. In particular we should take into account the possible occurrence of self-trapping effect for  $\text{Yb}^{3+}$  ions. This effect is producing re-absorption of photons emitted by excited state  $\text{Yb}^{3+}$  ions by surrounding  $\text{Yb}^{3+}$  ions at the ground state, which in turn re-emit and an increased lifetime is thus recorded. Such phenomenon is impossible to fully suppress and has to be and further investigated [13].

Regarding Tm co-doped glasses, from a compared analysis of Tab. 2 the decrease of  $\text{Yb}^{3+} ^2\text{F}_{5/2}$  lifetime with increasing  $\text{Tm}^{3+}$  concentration (group II glasses) is clearly showing that energy transfer from  $\text{Yb}^{3+}$  to  $\text{Tm}^{3+}$  is taking place. Besides it is interesting to note in Tab. 2 that, both for pump wavelengths of 980 and 785 nm,  $\text{Tm}^{3+} ^3\text{F}_4$  measured lifetime is decreasing with increasing Tm concentration (group II), whilst for group III with increasing Yb concentration this phenomenon does not occur. This is an evidence of the fact that  $\text{Yb}^{3+}$  ions are acting efficiently as sensitizers.

In particular, by comparing  $\text{Yb } ^2\text{F}_{5/2}$  lifetime in Y1 and T1Y1 samples we see that the energy transfer even for low Tm concentration has a time constant lower than the  $\text{Yb } ^2\text{F}_{5/2}$  lifetime. Therefore pump excitation of Yb is likely to be efficiently transferred to Tm ions. This is shown even better for higher Tm concentration in samples TxY1. In addition since the Yb concentration quenching is far larger than Tm concentration quenching we can conclude we will be able to effectively use Yb as sensitizer even with high Tm concentration and yet most of Yb excitation will be transferred to Tm and not lost radiatively.

Since in a previous publication [7] we had considered the optimum  $\text{Tm}^{3+}$  ions concentration between 4 and 5 mol% and since the  $\text{Yb}^{3+}$  ions concentration quenching is very high (13 mol%), we expect to be able to use an Yb:Tm ratio up to 3:1, even for  $\text{Tm}^{3+}$  concentrations up to 5 mol% to use Yb:Tm higher than 1:1, even for very high Tm concentration. In any case in fact we showed that energy transfer from Yb to Tm is very efficient even for Tm concentration as low as 1%. Future experiments will be however required to demonstrated such assumptions.

The fact that  $\text{Tm}^{3+} \ ^3\text{H}_4$  lifetime decays were not collected by pumping at 785 nm, except for T1Y1 glass, is probably due to the fact that pumping at 980 nm provides higher inversion than pumping at 785 nm.

A comparison of the lifetime values of group II glasses by pumping at 785 and at 980 nm with those collected in our previous work on Tm-only doped glasses was carried out and data are reported in Fig. 12. It is interesting to note that the values are in good agreement and that the presence of Yb does not affect the performance of the Tm ions, either by pumping at 785 nm or at 980 nm. The  $\text{Tm}^{3+} \ ^3\text{F}_4$  lifetime values, if compared to literature data, show a value of 0.65 ms for a total RE concentration of 5 mol% (4 mol% Tm and 1 mol% Yb) , which is the highest reported value up to now for such high rare earth concentrations [10; 11]. That is probably due to the fabrication process that in our case allowed minimization of OH content, an important source of quenching.

## 5. Conclusion

This paper shows that highly doped Tm-Yb tellurite glasses are feasible. This work also shows that Yb can be efficiently used as sensitizer of Tm ions to allow using cheaper and powerful diode pumping at 980 nm instead of 785 nm. We showed that Yb quenching concentration is of the order of 13% and far larger than Tm quenching concentration. This allows, if necessary, to use a Yb:Tm ratio up to 3:1, even for very high Tm concentration. We also showed that energy transfer from Yb to Tm is very efficient event for Tm concentration as low as 1 mol%. We therefore believe that our glasses are very promising host to develop efficient 980-nm pumped short cavity fiber laser. In particular the glass containing 4 mol% of  $\text{Tm}^{3+}$  and 1 mol% of  $\text{Yb}^{3+}$  is providing good spectroscopic properties and may be used to fabricate a short cavity optical fiber laser.

This work shows that different pumping schemes for  $\text{Tm}^{3+}$  are possible in order to obtain emission at 1820 nm without drastic decrease of quantum efficiency.

Future work will deal with the assessment of the Yb to Tm energy transfer coefficient in the presence of self trapping of Yb and the characterization of the visible emission of the samples in order to have a complete picture of the decay processes involved.

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## Figures

Fig. 1- Scheme of energy levels of  $\text{Tm}^{3+}$  and  $\text{Yb}^{3+}$  ions in a typical tellurite glass host.

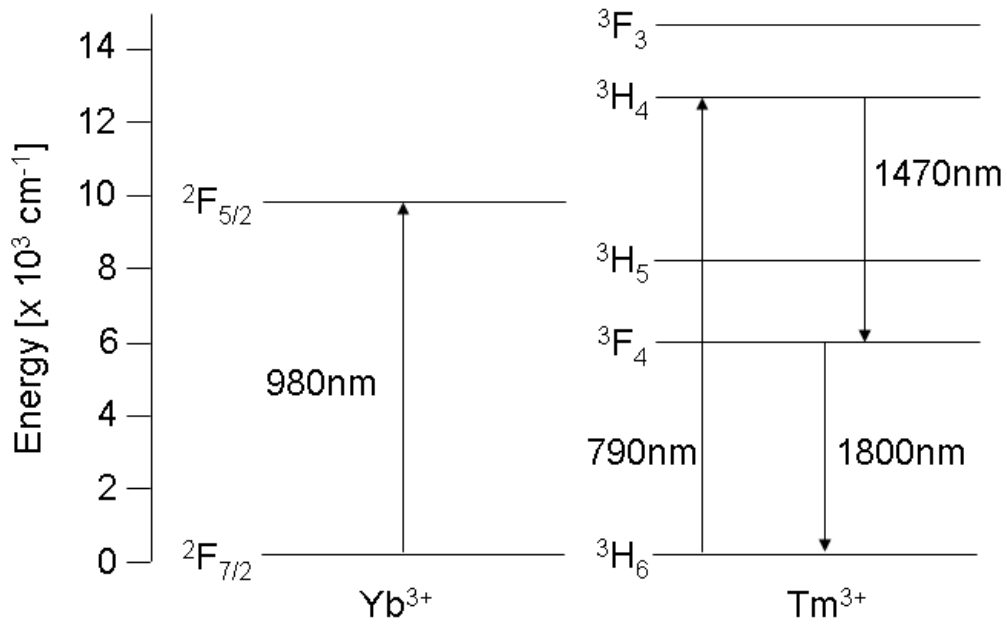


Fig. 2 - Refractive index measured values (at  $\lambda = 1533$  nm) of Tm-doped tellurite glasses as a function of  $\text{Tm}^{3+}$  ion concentration. Glasses displayed are: Tm-only doped tellurite glasses [7] as empty circles, group II glasses ( $\text{T}_x\text{Y}_1$  glasses where  $x = \text{mol\% Tm}^{3+}$  ion) and group III glasses (T4Y1, T4Y3 and T4Y5 glasses).

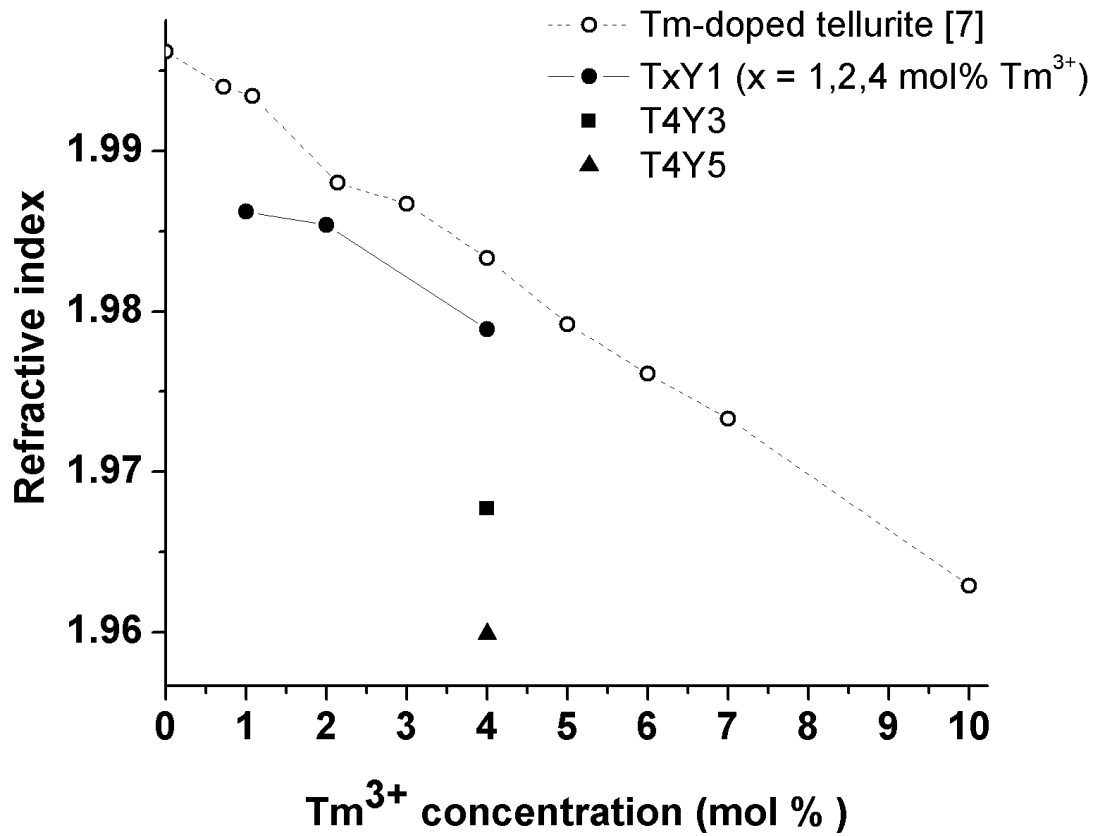


Fig. 3 – Example of the UV-VIS-NIR spectra of a Yb (Y5) and Yb/Tm (T4Y3) doped glasses. Each peak is labeled referring to the absorption transition from the ground state of  $\text{Yb}^{3+}$  in the case of Y5 glass and of  $\text{Tm}^{3+}$  in the case of T4Y3 glass (except for the  ${}^2\text{F}_{5/2}$  peak of this last glass, which refers to the  ${}^2\text{F}_{7/2} \rightarrow {}^2\text{F}_{5/2}$  of  $\text{Yb}^{3+}$ ).

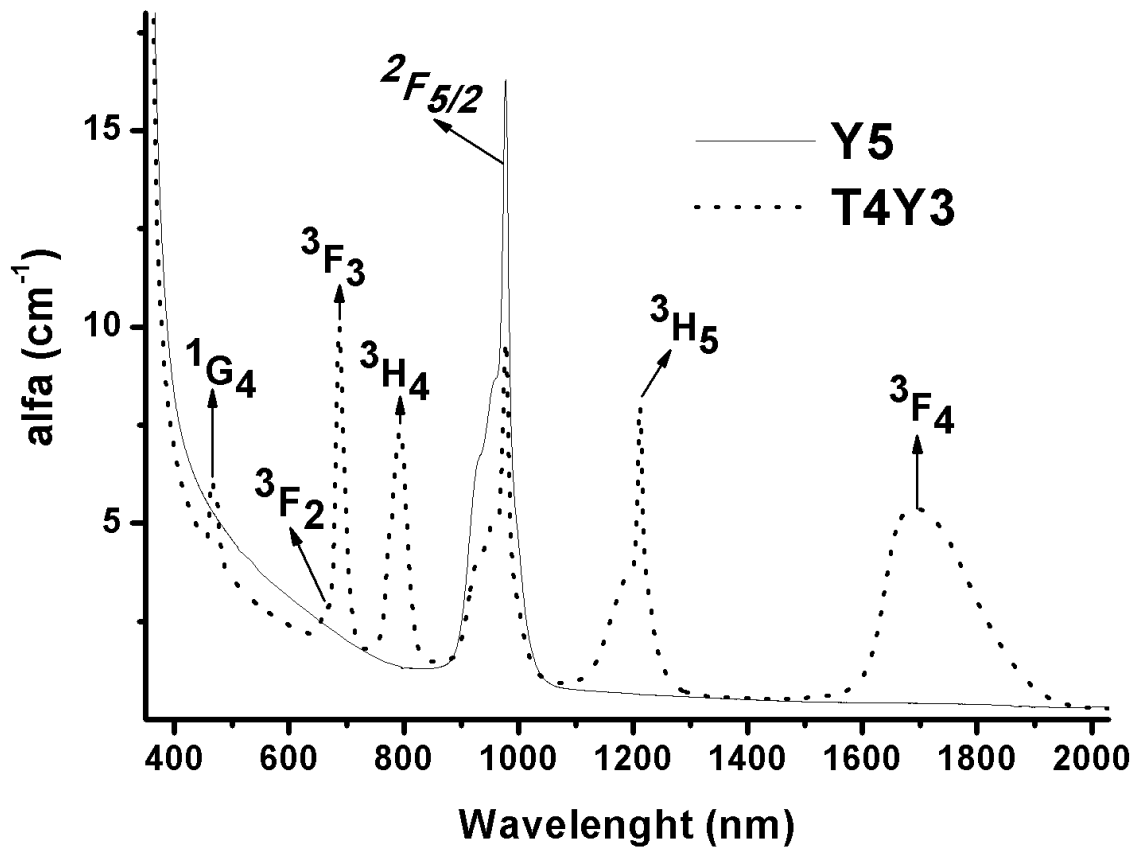


Fig. 4 - Absorption cross section of  $\text{Yb}^{3+} \ ^2F_{5/2}$  in Y5 glass (group I).

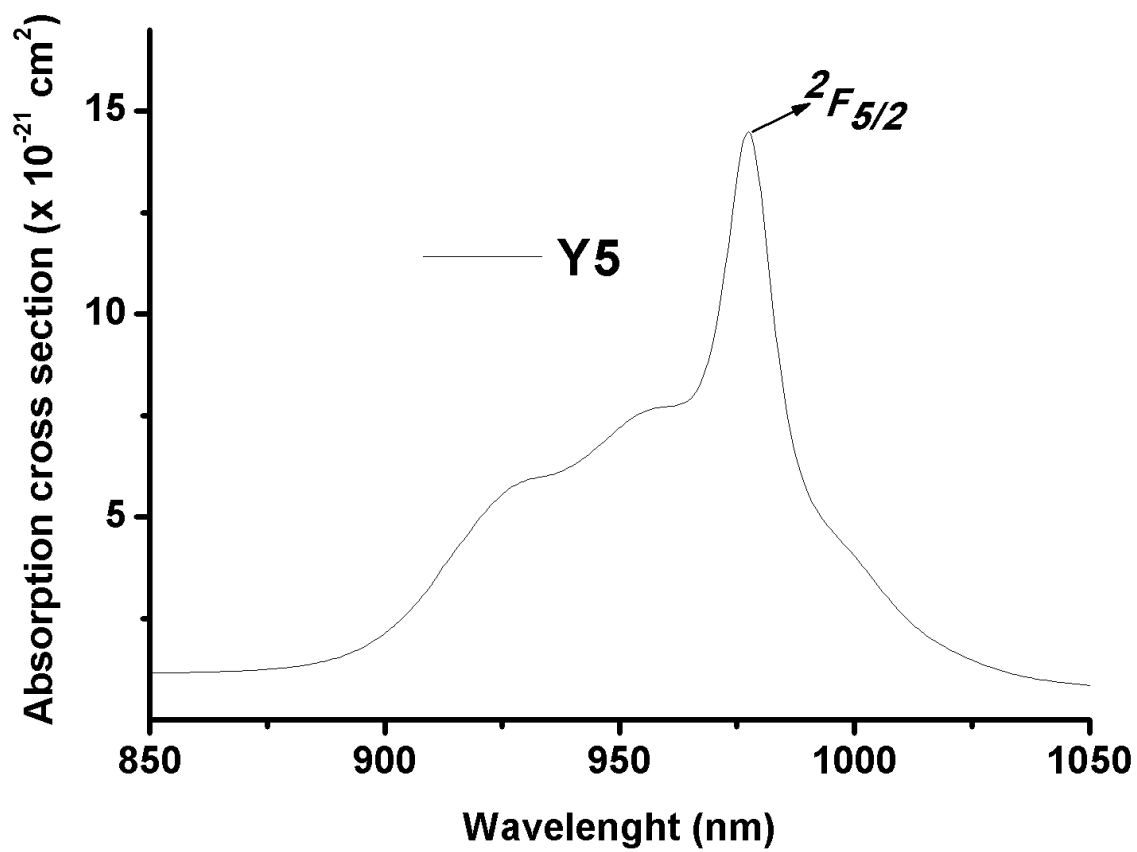




Fig. 5 - Fluorescence spectra of group II glasses (TxY1 glasses where x = mol%  $\text{Tm}^{3+}$  ion) by pumping at 980 nm.

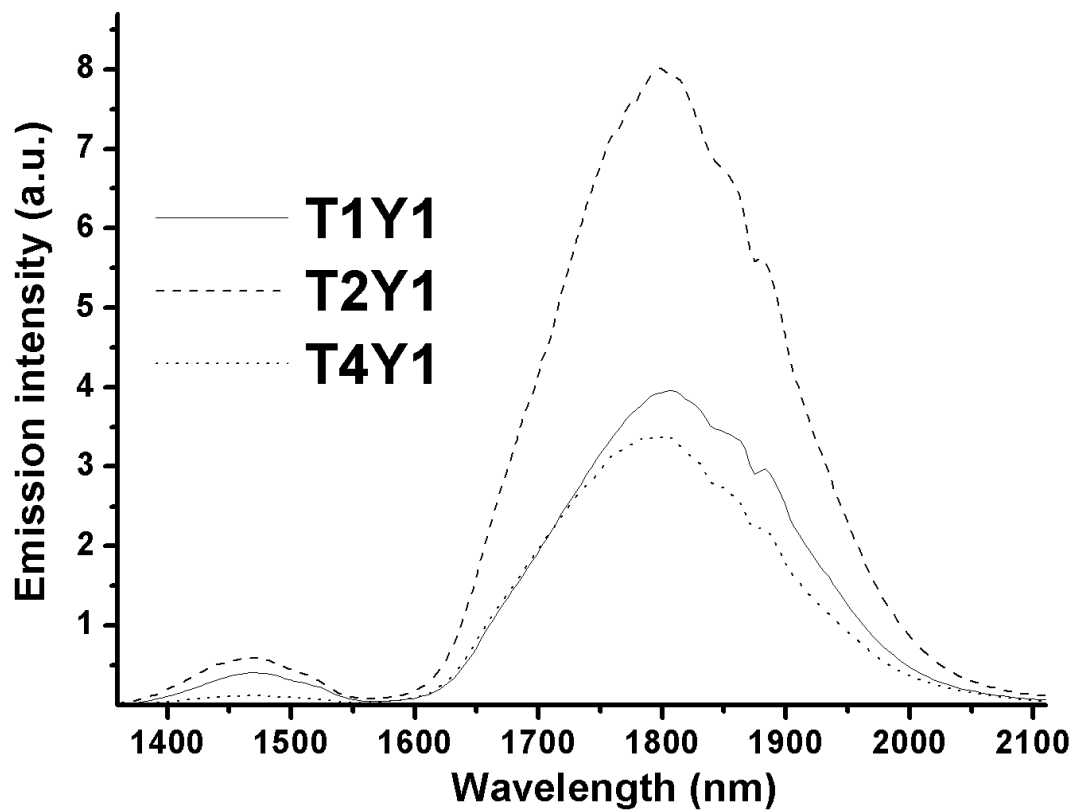


Fig. 6 – Fluorescence spectra of group III glasses (T1Yx glasses where x = mol% Yb<sup>3+</sup> ion), by pumping at 980nm.

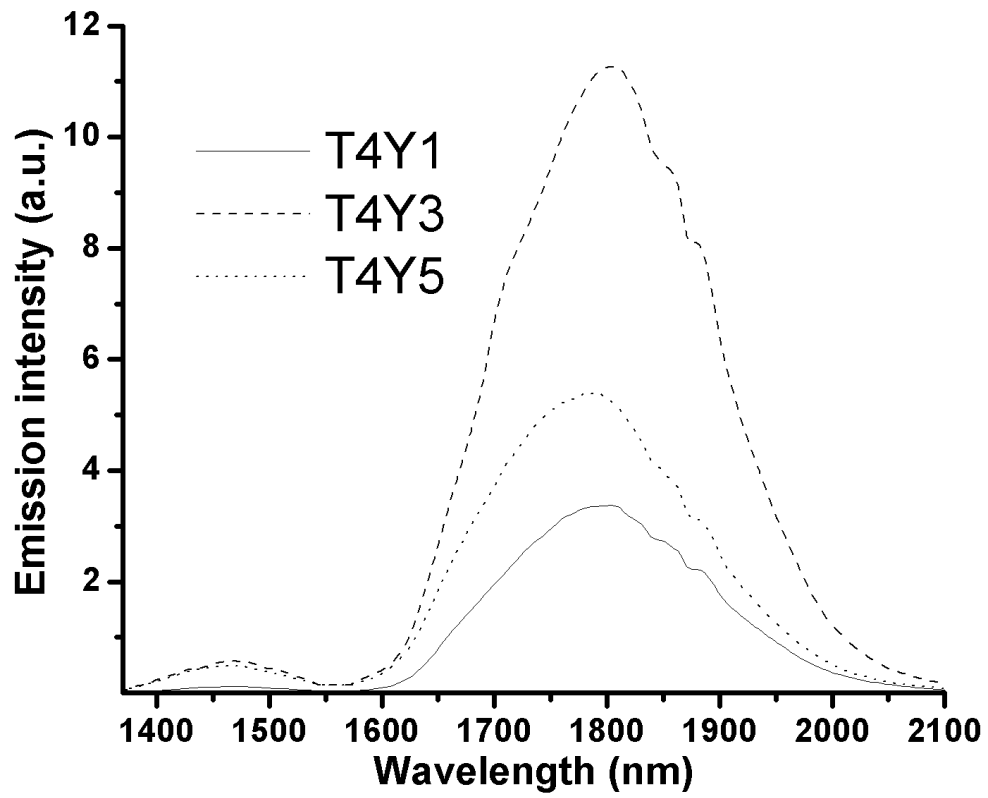


Fig. 7 - Intensity ratio between  ${}^3F_4 \rightarrow {}^3H_6$  and  ${}^3H_4 \rightarrow {}^3F_4$  emission transitions vs.  $Tm^{3+}$  concentration for TxY1 glasses (x = 1, 2, 4 mol% of  $Tm^{3+}$ ) by pumping at 980 nm.

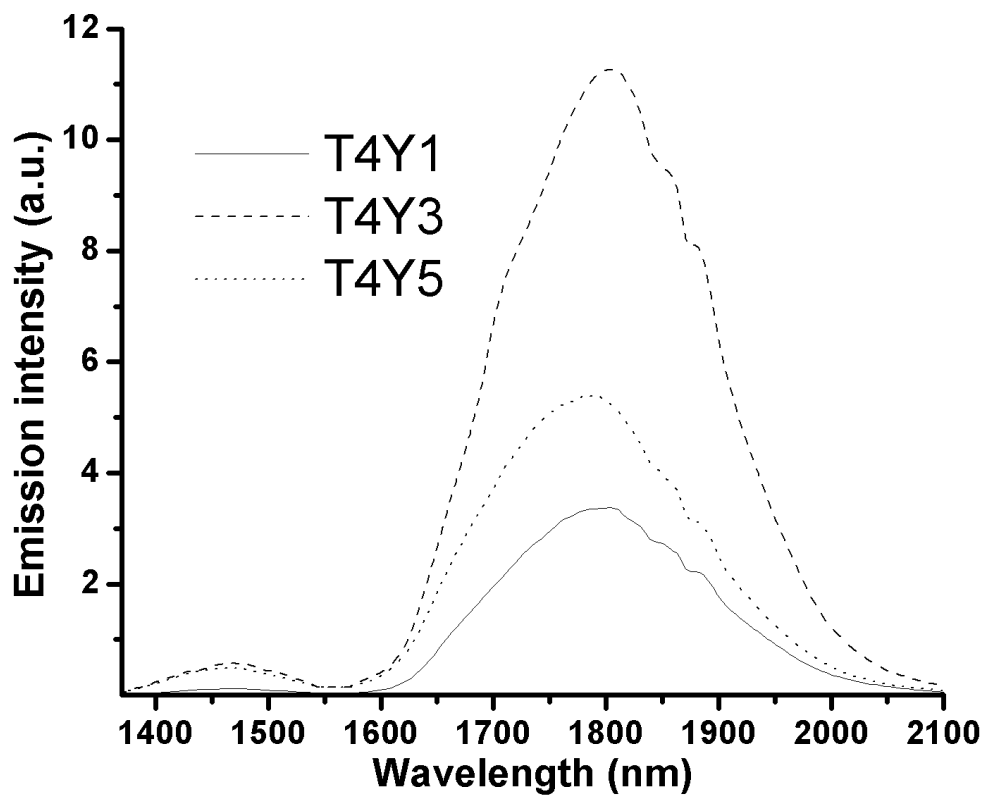


Fig. 8. Intensity ratio between  ${}^3F_4 \rightarrow {}^3H_6$  and  ${}^3H_4 \rightarrow {}^3F_4$  emission transitions vs.  $Yb^{3+}$  concentration for group III glasses (T4 Y<sub>x</sub>, x = 1, 3, 5 mol%  $Yb^{3+}$  ion) by pumping at 980 nm.

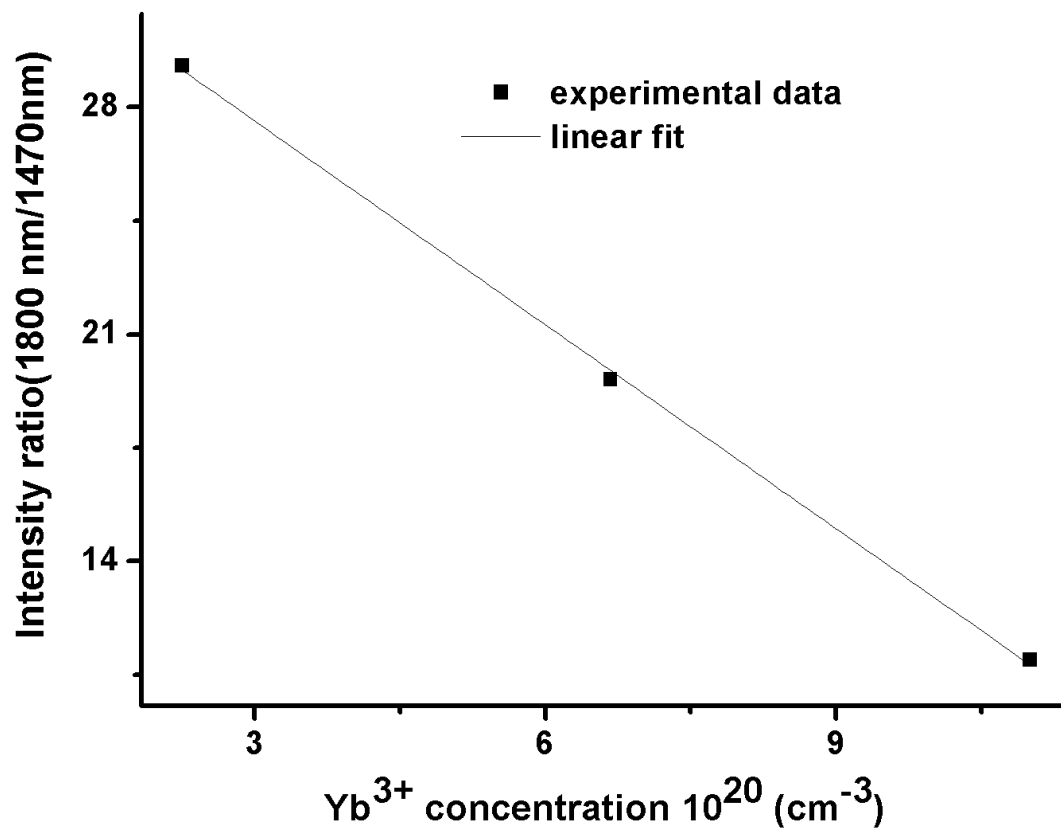


Figure 9 – Yb  $^2F_{5/2}$  lifetime decay curves by pumping at 980 nm.

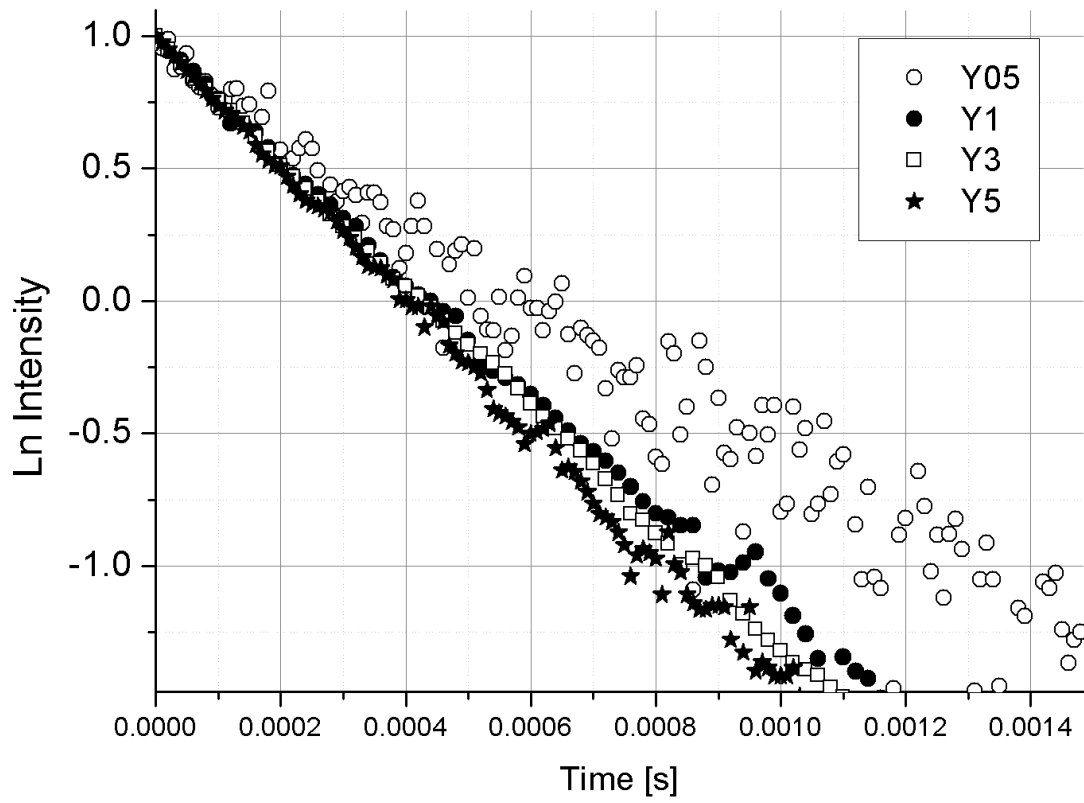


Fig. 10 -  $Tm^{3+}F_4$  lifetime for group II glasses by pumping at 785 nm.

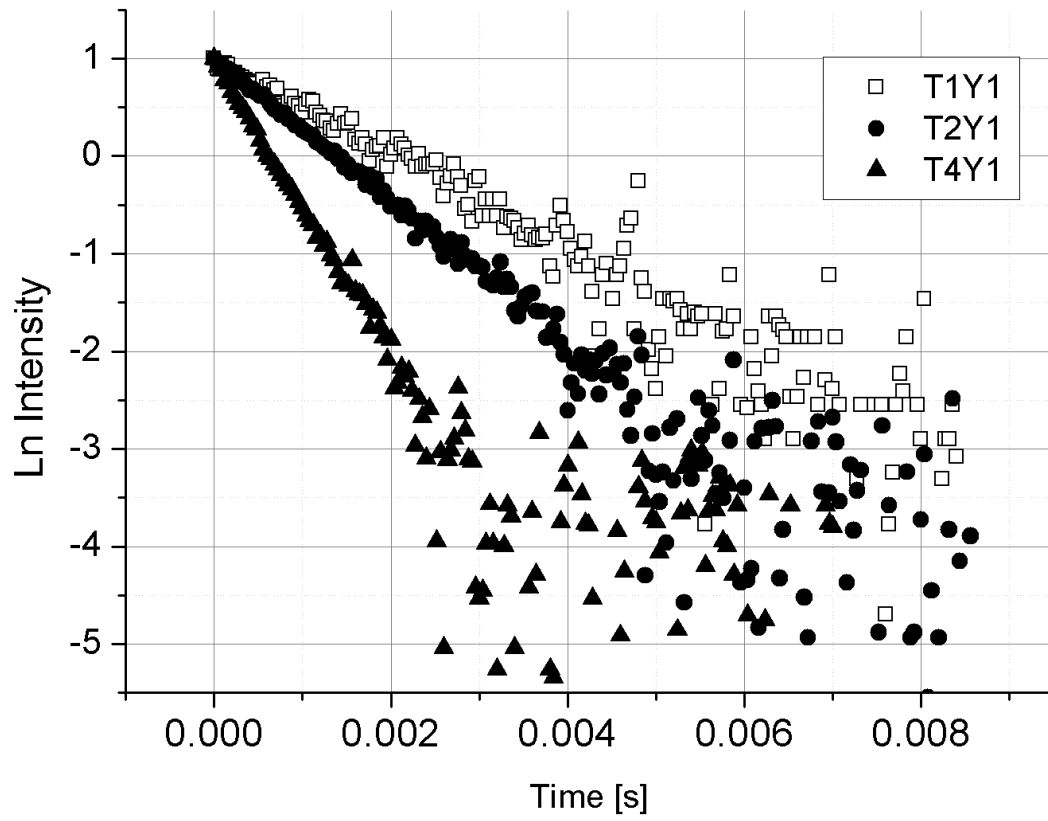


Fig. 11 – Fitting and experimental values of Yb lifetime from group I glasses using equation (1).

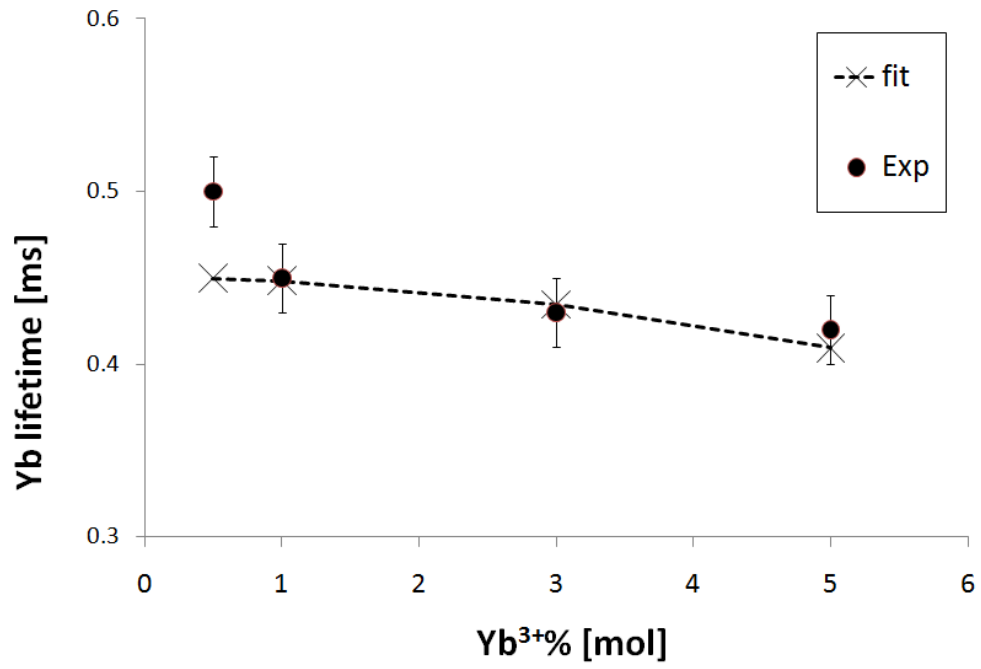
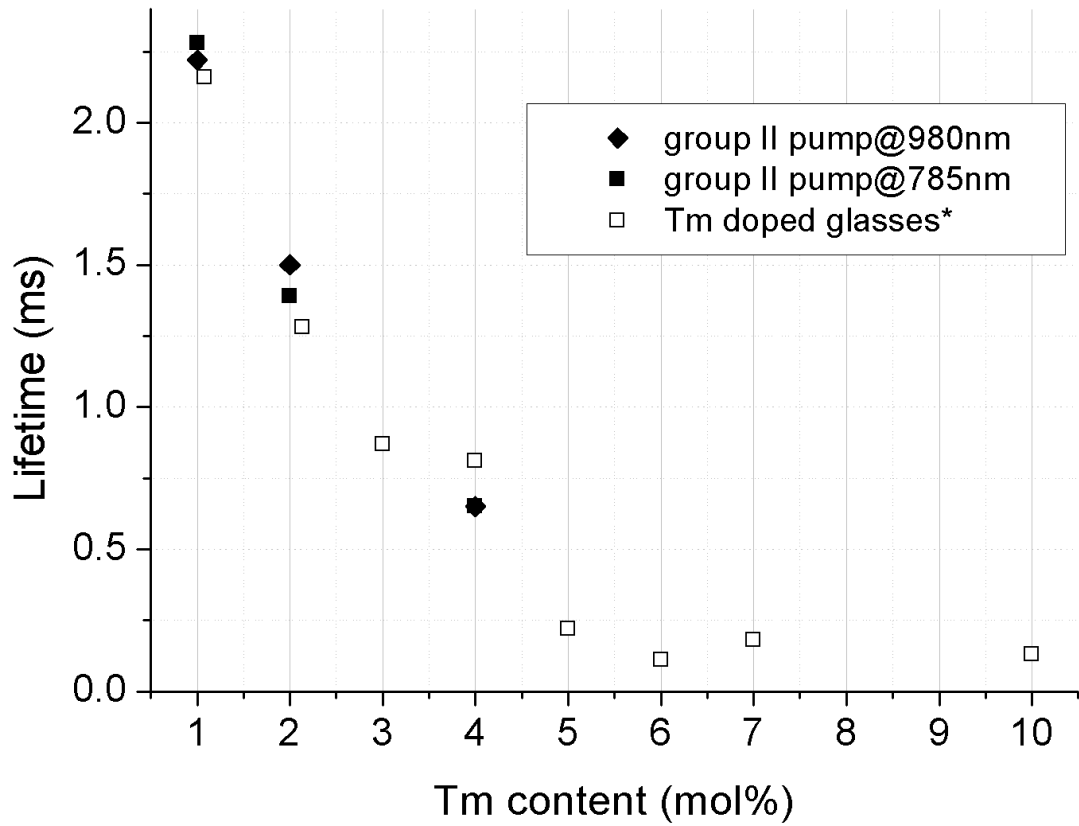


Fig. 12 - Comparison of Tm  $^3F_4$  lifetime for group II glasses and Tm doped glasses (previous paper submitted to J Non Cryst. Sol.). Tm doped glasses were pumped at 785 nm.





## Tables

Table 1 – Rare earth doped tellurite glasses prepared together with their main characteristic parameters: rare earth ion concentrations, density,  $T_g$ ,  $T_x$  and refractive index values.

<i>Group</i>	<i>Sample name</i>	$Tm^{3+}$ (ppm)	$Tm^{3+}$ ( $\times 10^{20}$ ions/cm <sup>3</sup> )	$Yb^{3+}$ (ppm)	$Yb^{3+}$ ( $\times 10^{20}$ ions/cm <sup>3</sup> )	$\rho$ (g/cm <sup>3</sup> )
<b>I</b>	<b>Y0.5</b>	0	0	6193	1.15	5.32
	<b>Y1</b>	0	0	12329	2.29	5.34
	<b>Y3</b>	0	0	36328	6.79	5.37
	<b>Y5</b>	0	0	59485	11.24	5.43
<b>II</b>	<b>T1Y1</b>	11932	2.27	11649	2.27	5.33
	<b>T2Y1</b>	23658	4.53	11548	2.26	5.37
	<b>T4Y1</b>	46512	9	11352	2.25	5.43
<b>III</b>	<b>T4Y1</b>	46512	9	11352	2.25	5.43
	<b>T4Y3</b>	45709	8.88	33469	6.67	5.45
	<b>T4Y5</b>	44934	8.83	54837	11	5.51

<i>Group</i>	<i>Sample name</i>	$T_g$	$T_x$	$T_g-T_x$	$n@825\text{ nm}$	$n@1061\text{ nm}$	$n@1533\text{ nm}$
<b>I</b>	<b>Y0.5</b>	312	440	128	2.0219	2.0073	1.9941
	<b>Y1</b>	312	442	130	2.0182	2.0036	1.9909
	<b>Y3</b>	321	474	153	2.0103	1.9960	1.9835
	<b>Y5</b>	327	480	153	2.0006	1.9871	1.9748
<b>II</b>	<b>T1Y1</b>	316	464	148	2.0130	1.9986	1.9861
	<b>T2Y1</b>	319	468	149	2.0211	1.9981	1.9854
	<b>T4Y1</b>	328	480	152	2.0053	1.9907	1.9789
<b>III</b>	<b>T4Y1</b>	328	480	152	2.0053	1.9907	1.9789
	<b>T4Y3</b>	333	478	145	1.9938	1.9803	1.9677
	<b>T4Y5</b>	337	460	123	1.9846	1.9718	1.9599

Table 2 – Lifetime values of all prepared glasses by pumping at 980 nm and at 785nm. All values are in ms and error is estimated around  $\pm 0.02$  ms.

Group	Sample name	Lifetime Yb <sup>3+</sup> (pump @980 nm)	Lifetime Tm <sup>3+</sup> (pump @980 nm)		Lifetime Tm <sup>3+</sup> (pump @785 nm)	
		<sup>2</sup> F <sub>5/2</sub>	<sup>3</sup> H <sub>4</sub>	<sup>3</sup> F <sub>4</sub>	<sup>3</sup> H <sub>4</sub>	<sup>3</sup> F <sub>4</sub>
I	Y0.5	0.50	-	-	-	-
	Y1	0.45	-	-	-	-
	Y3	0.43	-	-	-	-
	Y5	0.42	-	-	-	-
II	T1Y1	0.21	0.20	2.22	0.27	2.28
	T2Y1	0.16	0.08	1.5	n.a.	1.39
	T4Y1	0.10	0.05	0.65	n.a.	0.65
III	T4Y1	0.10	0.05	0.65	n.a.	0.65
	T4Y3	0.09	0.048	0.55	n.a.	0.56
	T4Y5	0.07	n.a.	0.65	n.a.	0.75