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Color tuning of YAG:Ce^{3+} phosphor-in-glass nanocomposites using a Sm^{3+} doped tellurite glass.

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White-light-emitting diodes usually combine a blue-LED based on indium gallium nitride and a cerium-doped yttrium aluminum garnet (YAG:Ce^{3+}) phosphor that presents light emission in the green-yellow [1]. The phosphors emission spectra doped with rare earth ions (REI) can be adjusted by appropriately modifying the compounds of the host lattice [2]. Furthermore, they are encapsulated in a silicone resin that eventually is degraded shortening the LED lifetime. An alternative to the organic encapsulating materials is to incorporate the phosphors into a glass matrix (PiG), which are more resistant to thermal degradation and are chemically stable [3-4]. In this work, a ZnO-GeO_2-TeO_2:Sm^{3+} (TGZ:Sm) glass and YAG:Ce^{3+} nanoparticles (YAG NPs) PiGs nanocomposites were fabricated, and the change in the emission of the PiGs with the variation in Sm^{3+} ions concentration in the glass matrix evaluated.

YAG NPs were synthesized by sol-gel method, in which nitrates of Al, Y and Ce were dissolved together with hexamine in an appropriate ratio, and the solution was then heated at 90°C for 24 h in a conventional oven for the reaction to proceed. The obtained gel was then dried at 200°C followed by calcination at 1030°C. A TGZ glass doped with 0.5 and 1 % mol of Sm^{3+} was used as glass matrix to incorporate YAG NPs, samples were labeled as PiG-1 and PiG-2. The TGZ glasses were fabricated as reported elsewhere [5]. The glasses were grinded and milled and then mixed thoroughly with YAG NPs in a mass ratio of 95/5 (glass/YAG). Afterwards, the powder mixtures were compacted in a press and heat treated at 500°C for 30 min. Microstructure and composition (EDS analysis) of the composites were analyzed in a JEOL-JSM-7800F microscope. Photoluminescence was monitored in a spectrofluorometer equipped with a 450 W ozone free Xe lamp, a Horiba Triax 320 excitation and an emission iHR320 monochromator.

As synthesized YAG NPs have sizes below 100 nm (Fig.1a), and after PiG processing the NPs are homogeneously distributed in the nanocomposite as it can be seen in Fig 1b). EDS analysis of sample PiG-1 (Fig. 1c) shows the presence of all expected elements that constitute nanocomposite. Fig. 2a) shows the excitation (PLE) and emission (PL) spectra of the YAG:Ce^{3+} nanoparticles. PLE spectrum was acquired at a 526 nm emission, two distinct excitation bands at 340 and 460 nm characteristic of YAG:Ce^{3+} phosphors were observed [1]. Fig.2b) shows the normalized emission spectra of PiG-1 and PiG-2 acquired with a 340 nm excitation light. The spectra of both samples are very similar having a broad emission band centered at 526 nm from $^5D_1$ - $^2F_{5/2}$ energy transition of Ce^{3+} ions. Sm^{3+} ions characteristic emission peaks ($^4G_{5/2}\rightarrow{}^6H_{5/2}$, $^4G_{5/2}\rightarrow{}^6H_{7/2}$, $^4G_{5/2}\rightarrow{}^6H_{9/2}$ transitions) are seen as small peaks overlapped with the main emission band, and slightly increase its intensity for PIG-2 sample with higher Sm^{3+} concentration. The emission band at 450 nm does not appear for pure YAG NPs. Thus it is possible that after heat treatment structural changes in the glass matrix occurred and could give origin to the emission in the blue region. Furthermore, this band also presented overlapped peaks that are consistent with absorption transitions from the ground state $^6H_{5/2}$–($^4M_{17/2}$, $^4F_{5/2}$, $^6I_{13/2}$, $^4M_{15/2}$, $^4I_{11/2}$, $^4I_{9/2}$) of Sm^{3+} ions. Chromatic coordinates of all samples global emission were estimated and are depicted in

the CIE1931 chromaticity diagram in Fig. 2c). The obtained (x, y) values were: (0.36432, 0.57009) for YAG NPs, (0.2954, 0.395) and (0.3139, 0.4312) for PiG-1 and PiG-2, respectively. YAG NPs color emission is in the green-yellow region as for the nanocomposites; the chromatic coordinates move towards cold white-light region for the TGZ glass matrix with lowest Sm^{3+} concentration.

In summary, a $ZnO\text{-}GeO_2\text{-}TeO_2:Sm^{3+}$ glasses were successfully combined with $YAG:Ce^{3+}$ nanoparticles to obtain a phosphor-in-glass nanocomposite showing a global emission color in the white-light region. Thus, the obtained nanocomposites are good candidates for solid-state-lighting applications

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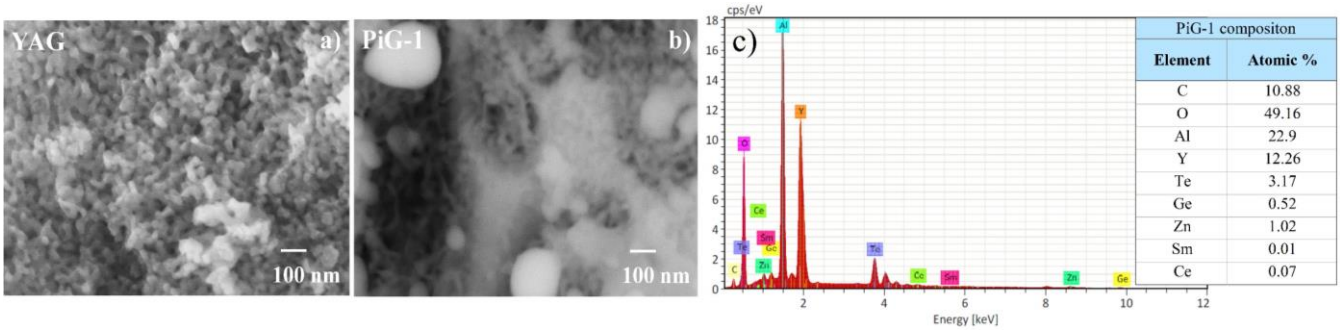

Fig.1. Micrographies of a) YAG NPs and b) PiG-1 were YAG NPs are incorporated into the glass matrix. c) EDS spectra and elemental composition of the PiG-1 sample.

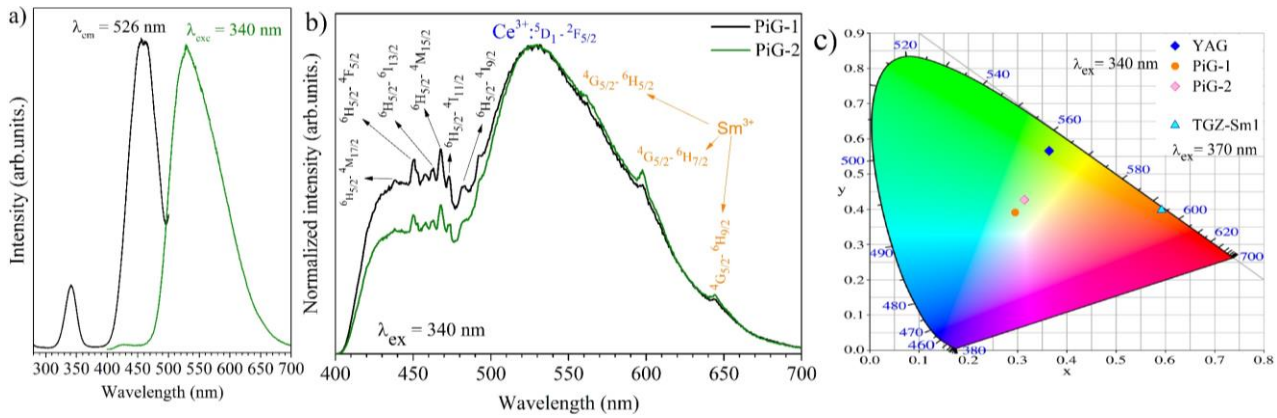

Fig.2. a) Excitation and emission spectra of the YAG. b) Normalized emission spectra of the PiG-1 and PiG-2 samples were recorded under 340 nm excitation. c) CIE chromaticity coordinates (x, y) of the YAG, PiG-1 and PiG-2 samples.

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