Upconversion and White Light Generation Properties of Undoped and Yb^{3+} doped Yttrium Silicate Nano-Phosphors

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ABSTRACT

In the present study, we have investigated the properties of yttrium silicate nanopowders undoped and doped with 5% and 10% (per mole) Yb^{3+}. The samples were obtained by the sol-gel method and annealed at 1250 ^{0}C for 12 hours. After the annealing process, the nanopowders were structurally characterized by X-Ray Diffraction (XRD) and is ascertained size was ~ 80 nm.

We have studied the emission from these samples when excited with the light of a near infrared laser diode at 975 nm.

Keywords: white light, up conversion, sol-gel method

1 INTRODUCTION

The optical systems that consist of rare earth ions embedded in a host lattice are very interesting materials for many applications such as optical sensing, data storage, nonlinear imaging and biomedical applications [1, 2, 3, 4]. Upconversion (UC) process generate one high energy photon out of two or more low energy photons. The transitions possible between different energy levels of a rare earth ion have spectral line shapes that described by the Dieke diagram [5]. Yttrium silicates (YSO) are suitable host materials for the rare earth (RE) ions because of their high thermal and chemical stability [6, 7].

The sol-gel method is used for preparing the Y_2O_3: SiO_2 nanocomposite since the technique has a lot of advantages such as needed a low temperature, minimizing the thermal decomposition, easy to control process. The devices and materials that are used in sol-gel technique are very simple. The needed temperature for processes without densification is low such as room temperature. The risk of thermal decomposition of the materials were minimized by this method and it is shown that high purity and stoichiometry are obtained by many experiments [6, 7, 8].

2 EXPERIMENTAL PART

The nanopowders of Y_2O_3-SiO_2 (YSO) undoped and doped with Yb^{3+} were synthesized by the sol–gel method and annealed at 1250 ^{0}C for 12 hours. The Yttrium(III) nitrate hexahydrate ($Y(NO_3)_3.6H_2O$- 99.9%), Ytterbium(III) nitrate pentahydrate ($Yb(NO_3)_3.5H_2O$- 99.9 %) salts and tetraethyl orthosilicate (TEOS, 99.9%) were purchased from Sigma – Aldrich. Also HCI, HNO_3, distillate water, ethanol were used for preparing the solutions. Ethanol and distillate water were used for solving $Y(NO_3)_3.6H_2O$ and $Yb(NO_3)_3.5H_2O$ with 1N 1cc HNO_3 as catalyst, separately. TEOS, distillate water and ethanol solution were prepared to initiate sol-gel reaction with 1N 1cc HCI as catalyzer at room temperature. After 20 minutes, $Y(NO_3)_3.6H_2O$ and $Yb(NO_3)_3.5H_2O$ solutions were added and mixed at 70 ^{0}C for 40 minutes. Then, TEOS, ethanol, distillate water and $Y(NO_3)_3.6H_2O$, $Yb(NO_3)_3.5H_2O$ solutions were mixed when the temperatures reached the room temperature. The solution were mixed for 2.5 h continously at room temperature. Finally, the samples were poured into petridishes and left in the dessicator at least 3-4 weeks to obtain glassy form. The samples were annealed at 1250 ^{0}C for 12 h and were grinded in agate mortar to produce nano-crystalline $Y_2Si_2O_7$ powders.

The optical characterization of the materials was performed by using a near infrared excitation at 975 nm with a diode laser LDI-820. The up conversion and white light emission spectra of the Yb^{3+} doped and undoped yttrium silicate nanopowders were measured between 450 nm and 900 nm by using a diode laser LDI-820 with 975 nm excitation wavelength and a McPearson Inc Model 2051 monochromator under 0.01 mbar pressure and different pumping power values. The slit was set at 500 micrometer for all samples and the optical signal was focused by EG&G Model 5210 lock-in amplifier and then detected by Hamamatsu R1387 photomultiplier tube. A short pass 900 nm cut off filter was placed between the excited sample and the monochromator's entrance slit.

The illuminescence meter results were obtained by using an Allied Scientific Pro ASP-MK350 Model

illuminator for determining the color quality parameters. All the measurements in this study were performed at room temperature. The room temperature decay and the rise time measurements for white light were carried out using a shutter to interrupt the laser beam and a Tektronix model TDS 3052B oscilloscope.

3 RESULTS AND DISCUSSION

The X-ray diffraction (XRD) images of the 5% per mole Yb^{3+} doped sample was taken by the Bruker D2 Phaser Model (Cu-Kα radiation) diffractometer setting in the 2θ range from 10^0 to 50^0 with scanning steps of 0.02.

Figure 1 shows the XRD patterns of the 5% Yb^{3+} doped sample annealed at 1250 ^{0}C for 12 h. The average particle size was estimated to be ~80 nm by the using Scherrer equation. According to JCPDS (Joint Committee for Powder Diffraction Data), the structure of the sample may be found mostly as α-$Y_2Si_2O_7$ with the card number 38-0223.

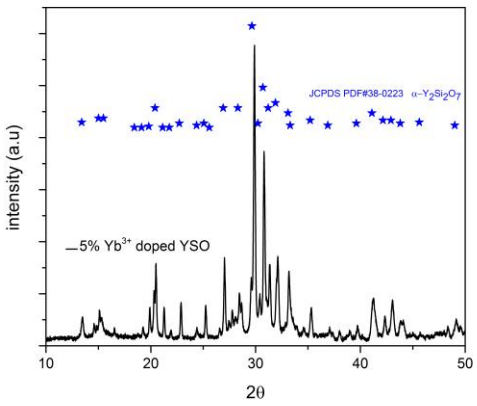

Figure 1. XRD patterns of the 5% Yb^{3+} doped yttrium silicate (YSO) nanopowders annealed at 1250 ^{0}C for 12 h

Luminescence spectroscopy is particularly apt at measuring peculiar aspects of nanopowders. We have observed up converted emission at ~ 475 nm, ~ 520 nm, ~ 653 nm, and ~ 710 nm due to f-f transition of Yb and unwanted Er impurities [6] by using 0.38 watt of laser power for 5% and 10% per mole Yb^{3+} doped yttrium silicate samples. Increasing the pumping power of the laser, the emission loses its detailed structure and eventually presents approximately the spectrum of white light.

Figure 2 shows the UC characteristics of 5% Yb^{3+} doped yttrium silicate sample. We observed the blue-green light emission at low power of 0.12 W – 0.92 W. For low pumping power part, blue green emission between 475 nm and 550 nm is due to cooperative emission of two Yb^{3+} ions and unwanted Er impurities.

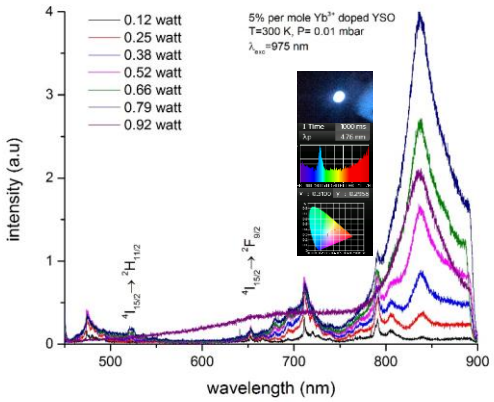

Figure 2. The blue-green emission and white light generation of 5% per mole Yb^{3+} doped YSO at 0.01 mbar pressure.

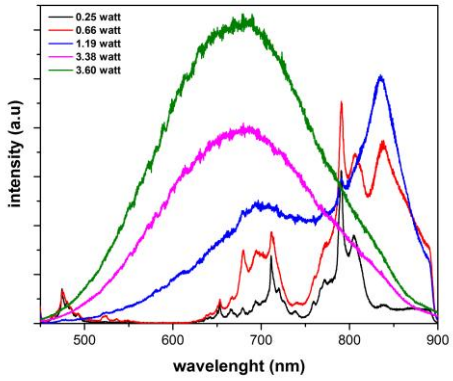

Figure 3. The white light generation of 10% per mole Yb^{3+} doped YSO at 0.01 mbar pressure.

Figure 3 shows the blue green spectra of 10% Yb^{3+} doped yttrium silicate sample at low pumping power and white light at higher power values at 0.01 mbar. The bright blue-green cooperative UC visible emission around ~ 475 nm in the spectrum is a cooperative emission corresponding to the simultaneous radiative relaxation of $Yb^{3+} \rightarrow Yb^{3+}$ ion pairs, green and red emission are $^4I_{15/2} \rightarrow (^2H_{11/2}, ^4S_{3/2})$ at ~521 nm and $^4I_{15/2} \rightarrow ^2F_{9/2}$ ~653 nm energy transitions of Er^{3+} impurities. The position of white light was ~700 nm for pumping power at 1.19 W. When the power was reached to 3.6 W, it was shifted to ~678 nm. The shape of the transition of Er^{3+} impurity is also changed with increasing power.

The white light emission was observed also for undoped sample (Figure 4). The white light of the undoped $Y_2Si_2O_7$ wasn't a strong as that of the other nanopowders. The peak position of the white light is ~745 nm below the 3.38 W.

Above 3.38 W, the maximum intensity is centered at ~716 nm with a broad range.

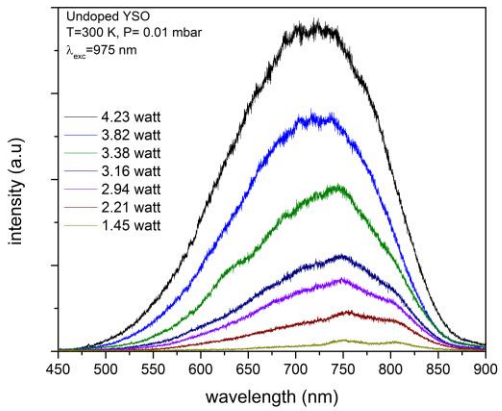

Figure 4. The white light generation of undoped YSO at 0.01 mbar pressure.

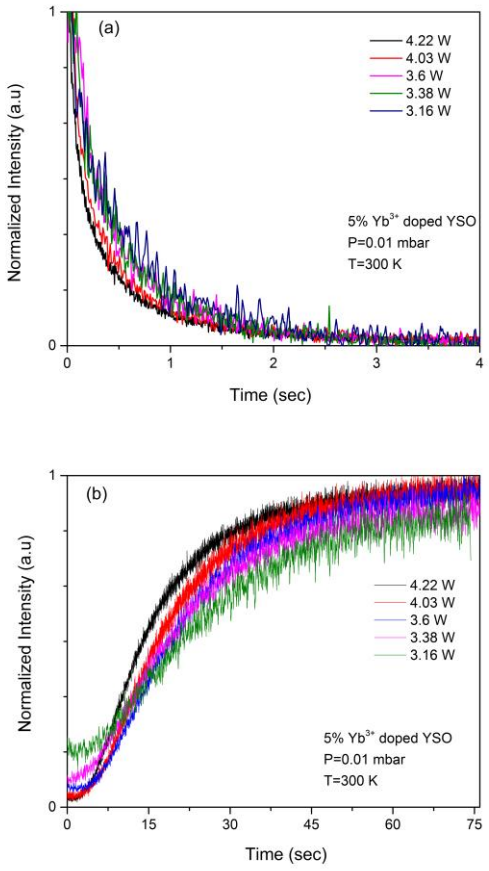

Figure 5. The decay (a) and rise (b) pattern of white light generation of 5% per mole Yb^{3+} doped YSO at 0.01 mbar pressure.

The decay patterns and rise patterns behaviors are shown in Figures 5a,b for 5% Yb^{3+} doped nanopowders. Figure 5a indicates that the decay patterns of 5% Yb^{3+} doped sample hasn't any strong dependence on pumping power; however, the rising patterns are sensitive to the pumping powers. As seen n Figure 5b, the rise pattern is fast at high powers and decreased to low power values gradually.

The chromaticity coordinates were found for all nanopowders samples in the white region of the 1931 Commission International I' Eclairage (CIE). The spectrum was measured by using the illuminance meter and the choromaticity coordinates of white light (0.4221,0.3726), (0.4673, 0.4073) and (0.3217, 0.2926) CIE 1931 for the samples 5%, 10% concentration of Yb^{3+} and undoped yttrium silicate nanopowders at 3.8 W under 0.01 mbar pressure.

4 CONCLUSION

In conclusion, 5% and 10% (per mole) Yb3+ doped and undoped yttrium silicate (YSO) nanopowders were synthesized using the sol-gel method. The blue-green UC emission and white light were obtained with various pumping power at 975 nm diode laser excitation at room temperature under 0.01 mbar pressure. The blue–green UC characteristics because of the cooperative emissions of two Yb^{3+} ions were obtained for the samples of 5% and 10% per mole Yb^{3+} doped yttrium silicate nanopowders at low pumping power values. When the pumping power was increased, the white light spectra was obtained for all samples. The experimental results show that the Yb^{3+} doped and undoped $Y_2Si_2O_7$ samples should be a promising material for novel white light sources for optical applications in many scientific and industrial area.

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