
ANALYSIS OF LUMINESCENCE MECHANISM OF HIGHLY EFFICIENT YAG AND TAG PHOSPHORS FOR LED's APPLICATION

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INTRODUCTION

The idea of a white light emitting diode (LED) seems not to be inherently unusual or surprising. It was not until the recent successful creation of high frequency light LEDs in the blue/ultraviolet region when the white LED made its debut. With the perspective of a highly efficient, cold light source, the LED market will have an enormous growth and the white LED will be a likely candidate for the replacement of the light bulb once production costs fall as the technology advances. The first commercially available white LED based on phosphors was produced by Nichia Co., which was also first to manage to make the blue LED. Nichia used a blue light emitting Gallium Indium Nitride and coated the chip with yellow fluorescent phosphor $Y_3Al_5O_{12}:Ce$, well known now as (YAG:Ce). A little later an other yellow phosphor $Tb_3Al_5O_{12}:Ce$ (TAG:Ce) was also used with GaInN chip for white light production. Usually these TAG:Ce and YAG:Ce phosphors are synthesized by solid state reaction (SSR) method and have the similar properties.

The aim of this study is to investigate the phosphors properties and luminescence mechanism of highly efficient YAG and TAG samples and propose how to improve and optimize the luminescence parameters.

EXPERIMENTAL DETAILS

The samples TAG:Ce and YAG:Ce as well as (Y, Tb)AG:Ce phosphors were synthesized by SSR in Korea Advanced Institute of Technology (KAIST) [1].

X-ray diffraction measurement – The crystal structures of the prepared samples were determined by X-ray diffraction measurement using goniometer (Rigaku, D/max- C(3kw) with $Cu-K\alpha$ ($\lambda = 1.5418 \text{ \AA}$) at 40 kV and 45 mA. The scan rate was 3 °/min and covered the range between 10° and 80°.

Photoluminescence (PL) measurement – In order to investigate the optical properties, PL was measured by using xenon lamp (500 W). Measurement was done at $\lambda_{exc} = 450 \text{ nm}$.

Morphology and size measurement – To observe the particle size and morphology, SEM micrographs of phosphor powders were taken by using SEM (Philips, XLFSEG).

RESULTS AND DISCUSSION

Figure 1 shows the PL spectra of TAG:Ce, (Y,Tb)AG:Ce, and YAG:Ce phosphor under 450 nm excitation. The maximum emission peak of TAG:Ce, (Y,Tb)AG:Ce, and YAG:Ce phosphor are at 552.5 nm, 545.5 nm, and 538nm, respectively. Theoretically, compared with YAG:Ce, the spectrum of the TAG:Ce is slightly shifted to the red, because of reduced local symmetry. The ionic radius of Tb^{3+} (1.18 Å) is a little bigger than Y^{3+} (1.16 Å). However, this shift is not sufficient to compensate for the lack of a red component. A broad yellow emission of YAG:Ce (as well as of TAG:Ce) phosphor is due to $5d \rightarrow 4f$ transition of Ce^{3+} ion.

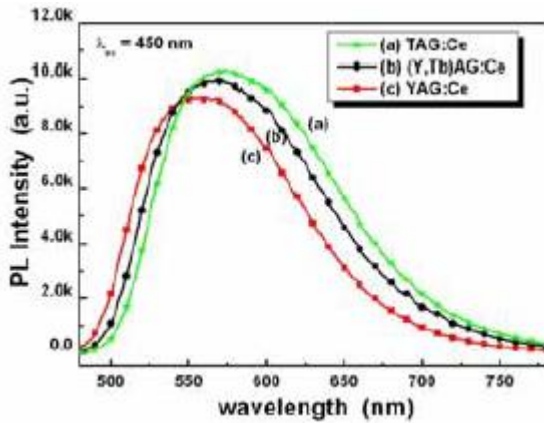


Fig.1. PL spectra of (a) TAG:Ce, (b) (Y,Tb)AG:Ce and (c) YAG:Ce phosphors

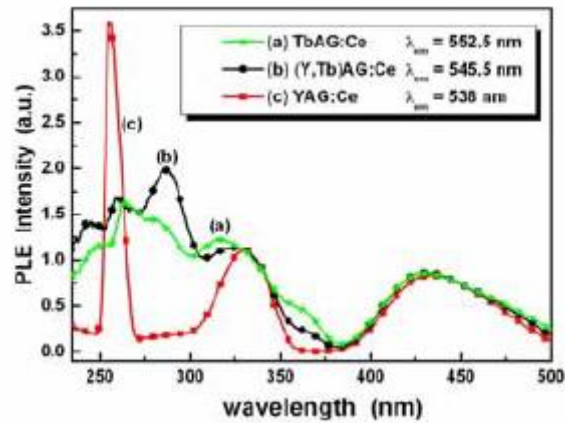


Fig. 2. PL excitation spectra of (a) TAG:Ce, (b) (Y, Tb)AG:Ce and (c) YAG:Ce phosphors

Fig. 2 shows the difference between PL excitation spectrum of TAG:Ce and that of YAG:Ce phosphor. The difference is remarkable in the ultraviolet region. Fortunately, in blue spectral region, two phosphors have the same excitation spectra, so that we can apply both these two phosphors to the white emitting light source using blue emitting LEDs. However, any improvement in the luminescence of these phosphors is extremely valuable to raise the light efficiency for different applications. To obtain white LED with high color rendering index (CRI), a separated red light source with secondary phosphor emitting at red region will work. Therefore, it is like to achieve the objective by co-doping Ce^{3+} and other rare earth ions into YAG and TAG phosphors. Red emission is found from a number of rare earth ion activators, such as trivalent Eu (at 590 and 610 nm), Sm (at 616 nm), or Pr (at 611 nm). All of them originate from $f-f$ transitions. Search is underway in finding suitable ions for co-doping with Ce to increase the red component to improve the CRI of white light. Using double Eu-Ce or Sm-Ce activation in YAG phosphor does not seem very fruitful, because it only diminishes the Ce luminescence, but using them in TAG could increase essentially the luminescence and improve CRI. In combination Eu-Tb-Ce terbium plays the role of sensitizer, it absorbs the exciting radiation from the LED and subsequently transfers this energy to Eu and Ce. Moreover, one additional way of transfer energy is possible: the radiation transitions in Tb may excite directly the neighboring Eu and Ce atoms. In such case Tb^{3+} in TAG does not serve as a doping element but becomes part of the host structure. The luminescence

mechanism could be understood from energy level diagram (Fig.3) and quantitatively is confirmed by 3-level model for energy transfer [2,3]

CONCLUSION

A codoping with Sm^{3+} , Pr^{3+} or Eu^{3+} looks very fruitful for improvement luminescence properties of YAG:Ce and TAG:Ce. The luminescence is more intense when the gap between the excited state and the more energetic component of the ground state multiplet, is larger. This explains the prominent role played by Eu^{3+} as co-activator. Introducing Eu^{3+} in the host lattice of TAG:Ce is more effective than in YAG:Ce. When Eu^{3+} is introduced in YAG, the part of exciting LED energy will excite the Eu^{3+} ions and reduce the Ce^{3+} excitation, that decrease the luminescence intensity. In the case of TAG, Tb^{3+} transfers its energy to Ce^{3+} and Eu^{3+} and can be used as a sensitizer. In addition, one new luminescent mechanism can exist when Tb luminescence excites direct Ce^{3+} and Eu^{3+} . The total luminescence increases and red shift appears, that improves CRI and CIE. It is very important to optimize the concentrations of activators and co-activator to improve the properties. TAG seems to be more perspective then YAG in the white light devices.

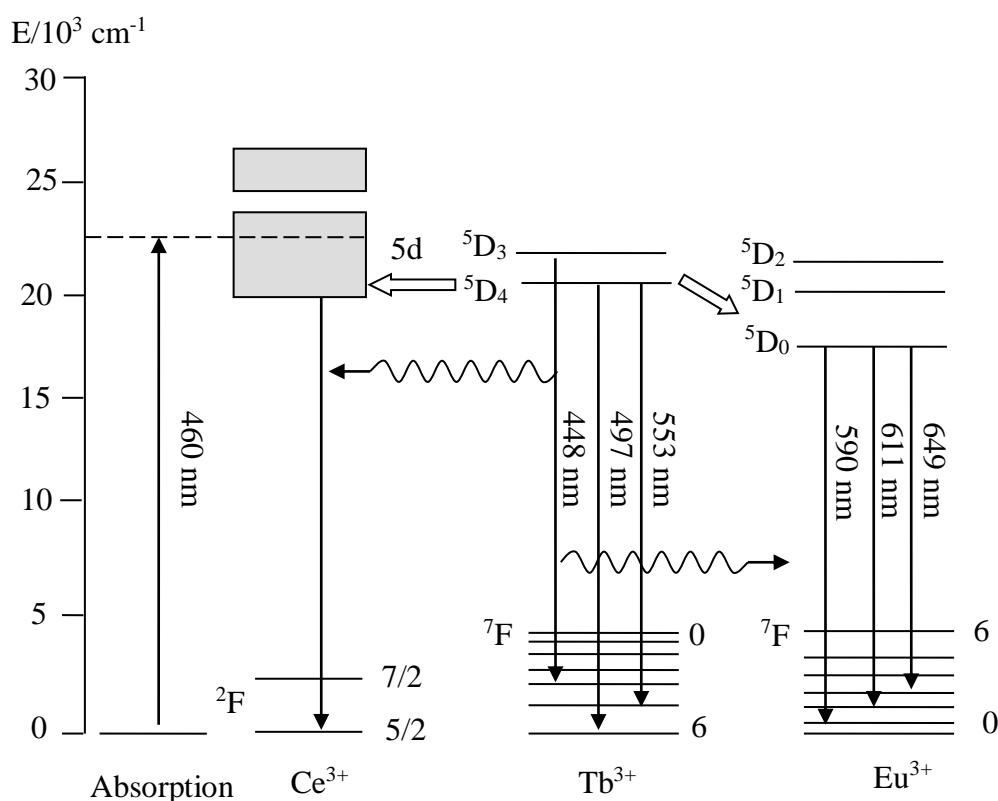


Fig.3. Luminescence mechanism in TAG:Eu,Ce

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