***Broad Band Visible Emission from Ce and Eu Co-doped in TAG Nano- phosphor***

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***Abstract* - Ce and Eu co-doped (dopent concentrations 0.1, 0.5 and 1.0mol%) in terbium alumina garnet (TAG) nano-powder was prepared by sol gel method followed by sintering at 1100oC using a home-built temperature controlled (±1oC) muffle furnace. The powder samples were characterized by XRD, SEM, FTIR and photoluminescence (PL) techniques. XRD and FTIR measurements showed that there was no change in the structure of TAG on doping SEM images showed that the powders contain mostly nano-sized spherical particles. TAG co doped with 0.1 mol% of Ce and Eu when excited by 280nm has broad band emission in 400-700nm region showing its potential for realization of white light source.**

***Keywords : TAG, Ce, Eu, sol gel, co doping, white light source***

I. Introduction

The inherently inefficient conventional light sources used for illumination are fast replaced by very efficient LED based white light sources world over resulting in enormous energy saving. An LED light source works on red-green-blue (RGB) color scheme where appropriate amounts of light from red-green and blue LEDs are mixed to produce white light. No doubt that these LED based white source are very energy efficient but their color rendering index (CRI) is inherently very low; CRI of a light source is its ability to reproduce the ‘true color” of the object illuminated by the light source[1]. Sunlight which is the best for reproduction of the color of an object faithfully has been assigned 100 as CRI while a filament lamp (CRI~100) is 2nd best for faithful color reproduction. On the other hand, the CRI of a commercial low cost LED based RGB white source mostly used for illumination can be as low as 28 while some very costly special sources with CRI of the order 90 have been developed for specific applications other than illumination.Hence, there is intense research activity in development of newer materials for white light sources materials which can match the energy efficiency of LEDs with CRI close to that of a filament lamp. This work is one such attempt where terbium aluminum garnet (TAG)nano-powders co-doped

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with rare-earth elements like Ce and Eu are investigated for their potential in the realization of white light sources with relatively higher CRI. Ce doped TAG is known to have strong emission (5d to 4f transitions) around 530nm [2] while Eu doped TAG emits around 600nm due to 4f to 4f transitions [3)]. This paper reports the synthesis and characterization of nano-powders of TAG co-doped with Ce and Eu to investigate the potential of this material for the realization of white light sources.

***II. Experimental detail***

Sol gel technique, as mentioned earlier,was employed for the preparation of both undoped and doped TAG nano-powders; terbium oxide (Tb4O7) and aluminum nitrate (Al(NO3)3.9H2O) being the starting materials for undoped TAG while cerium nitrate (Ce(NO3)3.6H2O) was used for doping Ce and europium oxide (Eu2O3) for Eu. The molar ratio of Tb4O7 to Al (NO3)3.9H2O used in the synthesis of undoped TAG was 3:5. Clear solution of Tb4O7 obtained by dissolving concentrated HNO3, was added to the aqueous (de-ionized water) solution of Al(NO3)3.9H2O. The mixture (mixture A) thus obtained was constantly stirred for about 3 hours maintaining the temperature at 65oC. This is followed by the addition of the citric acid (hydrated C6H8O7.H2O) solution which was prepared by dissolving about 0.04 moles of citric acid in distilled water. The whole mixture was maintained at 65oC and stirred continuously for about 10 hours to obtain the sol which turns into gel by heating over a water bath. The gel obtained was dried at 150oC in an oven. This dried gel was crushed into powder and sintered in air at 1100oC using a home-built temperature controlled (±1oC) muffle furnace. Same method is used for doped samples where appropriate amount of Eu2O3 solution prepared in concentrated HNO3 or aqueous solution (de-ionized water) of Ce(NO3)3.6H2O is added to mixture A and rest of the procedure as outlined above is followed exactly.

Powder XRD spectra at room temperature was recorded using Philips X perts x-ray spectrometer while SEM images were recorded on Joel JSM-7600F instrument. FTIR spectra were recorded using Shimadzu, FTIR 8000 series spectrometer. Photoluminescence was recorded using Carry 8000.

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| --- | --- | --- | --- |
| S.No | Host | Dopent (mol%) | |
| TAG | Ce | Eu |
| 1. | 0.5 | - |
| 2. | - | 0.5 |
| 3. | 0.1 | 0.1 |
| 4. | 0.1 | 0.5 |
| 5. | 0.1 | 1.0 |
| 6. | 0.5 | 0.1 |
| 7. | 0.5 | 0.5 |
| 8. | 1.0 | 0.1 |

**Table I. Co dopent concentration in TAG**

***III. Result and Discussion***

**Structural properties:** The structural properties as mentioned earlier were studied by XRD, FTIR and SEM techniques. Fig.1.shows the typical XRD pattern of TAG co-doped with Ce (0.1 mol. %) and Eu (0.5 and 1.0 mol. %) along with that of undoped TAG. All the three XRD spectra in Fig.1 look exactly same implying that co-doping does not change the structure of TAG. Similar results were obtained using the other co-doping combinations including 1.0 mol. % doping levels of both Ce and Eu. This conclusion is further supported by FTIR shown in FIG. 2, which compares the FTIR spectra of undoped TAG with those TAG co-doped with Ce (0.1 mol. %) and Eu (0.1, 0.5 and 1.0 mol. %). Similar results were obtained at the co-doping combinations. In Fig. 2 absorption peaks in lower frequency region at 502 cm-1,555 cm-1, 679 cm-1,710 cm-1,and 775 cm-1can be assigned to the metal oxide bonds (Ce-O, Tb-O ,Eu-O and Al-O). [4]

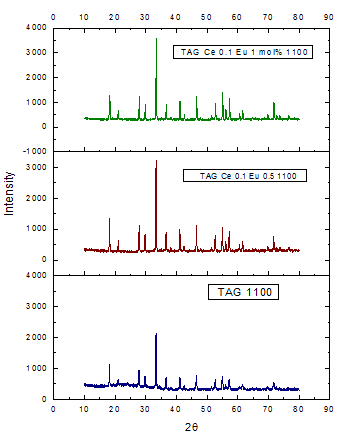
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Fig.1.XRD pattern of co doped and undoped TAG.

The particle size and morphology of sintered powder were examined by scanning electron microscope (SEM) and a typical SEM image is shown in Fig. 3. As seen in Fig. 3 the co-doped TAG powder contains nano-sized particles which are almost spherical in shape.Some bigger particles are due to agglomeration which is observed at high sintering temperatures. [5]

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Fig.3. SEM image of TAG: Ce 0.1 Eu 1 mol% at 1100­­o C

This investigation, as mentioned earlier, aims at the assessment of Ce and Eu doped TAG nano phosphorous as a material suitable for white light source which needs detailed study of room temperature photoluminescence in 400-700 nm regions. Photoluminescence spectra excited by 280nm radiation from a Xe lamp was recorded for various co-doped combinations as listed in Table I. Out of all these spectra Fig.5 and Fig.6 show an interesting result which indicate the potential of this material for the realization of white light source. As seen in this figure TAG doped with 0.1 mol.% of Ce and 0.5 mol. % Eu emits in a relatively broader band spanning from 400 nm to almost 700 nm indicating that by proper concentration tuning one might make the emission band flatter as well broader so that white light source based on TAG nano phosphorus co-doped with Eu and Ce can be realized. The exact mechanism responsible for this broadening is being investigated by concentration studies.

***IV. Conclusions***

The preliminary results of this investigation show that when TAG nano phosphorus is co-doped with 0.1 mol. % Ce and 0.5 mol. % Eu it emits from 400-700nm region when excited by 280nm radiation indicating the potential of this material for the realization of white light source. More investigation by changing the dopent concentration are required to understand the exact mechanism responsible for this broad emission.

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