ISSN: 0975-833X

Available online at http://www.journalcra.com

International Journal of Current Research
Vol. 5, Issue, 03, pp.529-531, March, 2013

INTERNATIONAL JOURNAL OF CURRENT RESEARCH

RESEARCH ARTICLE

SYNTHESIS AND UV, VUV PHOTOLUMINESCENCE OF RED EMITTING BORATE HOST PDP PHOSPHORS $YCaBO_4$: Eu^{3+} and YBO_3:Eu^{3+}

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ARTICLE INFO

Article History:
Received 27th December, 2012
Received in revised form
08th January, 2013
Accepted 21th February, 2013
Published online 19th March, 2013

Key words:
Inorganic Borates, Oxyborates, Combustion Synthesis, Red Emitting Phosphor, PDP, VUV, Hg-Free Fluorescent Lamp.

ABSTRACT

The Orange red and red emitting borate host phosphor $YCaBO_4$: Eu^{3+} and YBO_3: Eu^{3+} has been prepared by a novel solution combustion technique. The synthesis is based on the exothermic reaction between the fuel (Urea) and Oxidizer (Ammonium nitrate).The Heat evolved in reaction is utilized for auto combustion. The photoluminescence properties of the powder samples of $YCaBO_4$: Eu^{3+} and YBO_3: Eu^{3+} has been investigated under UV and VUV excitation. The phosphor shows strong absorption in UV and VUV region and exhibits intense red emission upon excited by 254 nm UV and 173 nm VUV radiation. Under UV 254 nm excitation, $YCaBO_4$: Eu^{3+} and YBO3:Eu^{3+} exhibits intense red emission around 610 nm. Under VUV excitation of 173 nm, the phosphor emits intense red emission around 610 nm and few weak emissions. These weak emissions could be suppressed by annealing the sample repeatedly at proper temperature and both the borate phosphor $YCaBO_4$: Eu^{3+} and YBO3:Eu^{3+}could be a good red emitting phosphor for PDP display applications and mercury free lamps.

INTRODUCTION

The plasma display panels (PDP) are gaining attention due to their high performance and scalability as a media for large format television (TV), particularly high definition TV (HDTV). Color plasma display panels (PDPs) have attracted considerable interest in recent years as components of wall-mounted television sets that are large, flat and thin (Bizarri, Moine, 2005; Rao, 2005; Zhang *et al*., 2005; Lin *et al*., 2007). The luminescence efficiency of a PDP depends upon various components such as phosphors, gas mixture, dielectric layer, reflective layer, black matrix, etc. The phosphor particles for PDP applications should have good luminescent characteristics under vacuum ultraviolet (VUV) light consisting of the resonance radiation of Xe atoms (147 nm) and the excited state of molecular Xe (172 nm). Recently, borates and oxyborates have been extensively investigated as host lattices for luminescent materials because of their large band gap and stability. Most borate hosts are transparent up to 140-180 nm, so that the VUV light can directly excite impurity activator in these hosts (Jung *et al*., 2005). The strong absorption due to the impurity ions would give efficient conversion of the VUV light. Several Inorganic Borate host materials have been reported as Red PDP phosphors, such as (Y, Gd) BO_3:Eu^{3+},$YAl_3(BO_3)_4$:Eu^{3+}, and BaZr $(BO_3)_2$:Eu^3, $YCa_4O(BO_3)_3$: Eu^{3+}, $GdAl_3(BO_3)_4$: Eu^{3+}, (Lee *et al*., 2002; Tian *et al*., 2004; Dorenbos and Lumin 2005; Yokosawa, Suzuki and Nakazawa 2003; Hongpeng *et al*., 2000; Park *et al*., 2001). The Rare earth Alkaline earth Metal Borate phosphor $YCaBO_4$: Eu^{3+} and YBO3:Eu^{3+} are interesting and useful l red phosphors. $YCaBO_4$: Eu^{3+} has been investigated in the recently years as a VUV excited phosphor for PDP applications. Park *et al.* (2001) investigated the possibilities of Ca(Y, Gd) (Al, Eu) BO_4 as red PDP phosphor (Park *et al*., 2008). In the present work we prepared the $YCaBO_4$: Eu^{3+} and YBO3:Eu^{3+} by a novel solution combustion technique and investigated the photo luminescent properties under UV and VUV excitation.

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Synthesis and Experimental Techniques

The samples were prepared by a novel technique which is slight variation of solution combustion technique (Sonekar *et al*., 2007; Sonekar *et al*., 2009; Sonekar, Omanwar and Moharil 2009; Nagpure *et al*., 2011). The starting ingredients Y $(NO_3)_3$ and Eu $(NO_3)_3$ (IR Ltd.), $Ca(NO_3)_2$, H_3BO_3, NH_4NO_3 and NH_2-CO-NH_2 (S D Fine AR) were used. The stoichiometric amounts of the ingredients (Table 1 and Table 2) were thoroughly mixed in an Agate Mortar, adding little amount of double distilled water and obtained an aqueous homogeneous solution. The aqueous solution was then transferred in to a china basin. The China basin was introduced in to preheated muffle furnace maintained at 550°C. The solution boils, foams and ignites to burn with flame and obtained a voluminous, foamy powder. The entire combustion process was over in about 5 minutes. Following the combustion, the resulting fine powders were annealed in a slightly reducing atmosphere provided by burning charcoal at temperature 800°C for about 60 min. and suddenly cooled to room temperature. PL measurements under UV excitation were carried out at NEERI, Nagpur on Hitachi F- 4000 Spectro-fluorometer and PL measurements under VUV excitation were carried out at Nanotechnology Application Centre, University of Allahabad, Allahabad (UP).

RESULT AND DISCUSSION

Figure 1 shows the UV Excitation and emission spectra of $YCaBO_4$: Eu^{3+}. The phosphor $YCaBO_4$: Eu^{3+} exhibits intense red emission under UV excitation. The excitation spectrum for 611 nm emission

Table 1.

$YCaBO_4:Eu^{3+}$ Molar ratio Weights (gms.)	$Ca(NO_3)_2$	$Y(NO_3)_3$	$Eu(NO_3)_3$	H_3BO_3	NH_2CONH_2	NH_4NO_3
	1	0.98	0.02	1	7	8
	2.3615	3.8299	0.0892	0.6183	4.2042	6.4032

Table 2.

$YBO_3:Eu^{3+}$ Molar ratio Weights (gms.)	$Y(NO_3)_3$	$Eu(NO_3)_3$	H_3BO_3	NH_2CONH_2	NH_4NO_3
	0.98	0.02	1	7	8
	3.8299	0.0892	0.6183	4.2042	6.4032

mainly consists of an intense double humped broad band peaking at 242–248 nm. The emission spectrum under 254 nm UV excitation mainly consists of a sharp intense line peaking at 610.4 nm which corresponds to the $^5D_0 \rightarrow {}^7F_2$ transition of Eu^{3+}. The other weak lines 589, 649 and 630 nm also occurs which corresponds to the $^5D_0 \rightarrow {}^7F_1$ and $^5D_0 \rightarrow {}^7F_3$ transition of Eu^{3+} ions. The prominent emission line 610.4 nm is due to the $^5D_0 \rightarrow {}^7F_2$ transition of Eu^{3+} which indicates that the Eu^{3+} ions occupies a noncentro-symmetric positions in the $YCaBO_4$ lattice. The red emission of $YCaBO_4$ is comparable to the commercial red phosphor $Y_2O_3:Eu^{3+}$. Hence it could be a potential red phosphor for lamp and display applications.

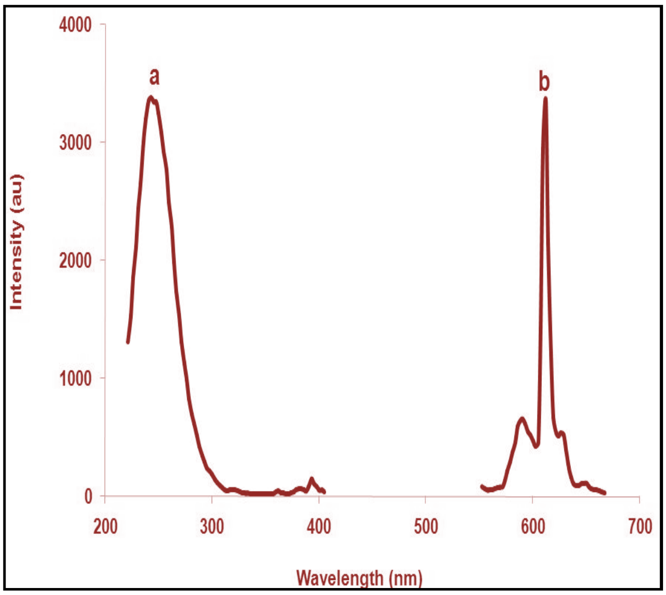

Figure 1. Excitation (a) for 611 nm emission and Emission Spectra for UV 254 nm excitation of $YCaBO_4$: Eu^{3+}

Figure 2 shows the emission spectrum of $YCaBO_4$: Eu^{3+} under VUV excitation of 171 nm. The emission spectrum under VUV excitation of 173 nm mainly consists of a sharp intense line peaking at 610.4 nm which corresponds to the $^5D_0 \rightarrow {}^7F_2$ transition of Eu^{3+}. The VUV emission spectrum shows some weakemissions. If these weak unwanted emissions are suppressed then the phosphor $YCaBO_4$: Eu^{3+} could be an excellent candidate for red emitting PDP phosphor. The $YBO_3:Eu^{3+}$ phosphor was prepared by combustion synthesis. The UV Photoluminescence spectra of $YBO_3:Eu^{3+}$ phosphor is depicted in Figure 3. The excitation spectrum (a) monitored at 594 nm emission shows a single intense broad band peaking at 240 nm. This band corresponds to the charge transfer (CT) transition within the Eu^{3+} - oxygen center. The emission spectrum (b) under the 254nm excitation, consists of multiple peaks at 594,612 and 627 nm which corresponds to the $^5D_0 \rightarrow {}^7F_1$, $^5D_0 \rightarrow {}^7F_2$ and $^5D_0 \rightarrow {}^7F_3$ transitions of Eu^{3+}. The prominent emission occurs at 594 nm in the orange red region of the spectrum. The red emission at 612 nm is comparatively weak. The VUV emission spectrum of $YBO_3:Eu^{3+}$ phosphor under 172 nm excitation is shown in Figure 4. The VUV emission spectrum is similar to the UV emission spectra. This clearly indicates that the phosphor exhibits strong absorption in the VUV region and efficient emission in the orange red region. The maxima observed at 596 nm and 611 nm in the red region of the spectrum are due to $^5D_0 \rightarrow {}^7F_1$ and $^5D_0 \rightarrow {}^7F_2$ transitions of Eu^{3+} ion. This is in good agreement with the literature value for the VUV excited luminescence.

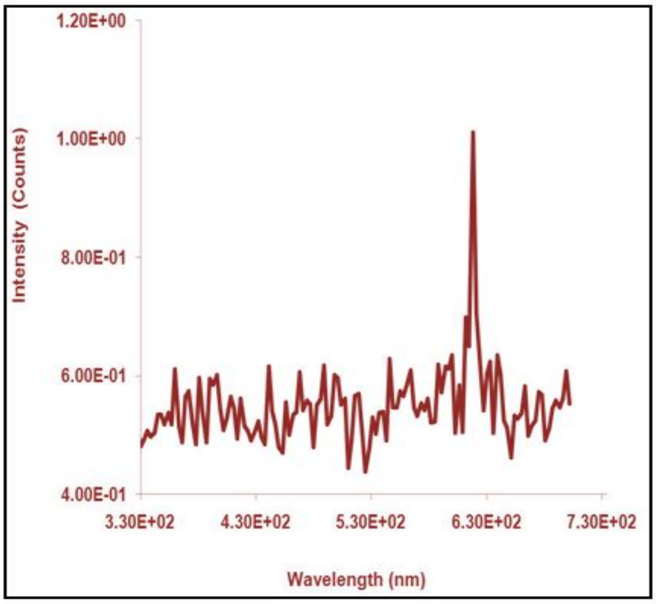

Figure 2. Emission spectra for VUV 172 nm of YCaBO4:Eu^{3+}

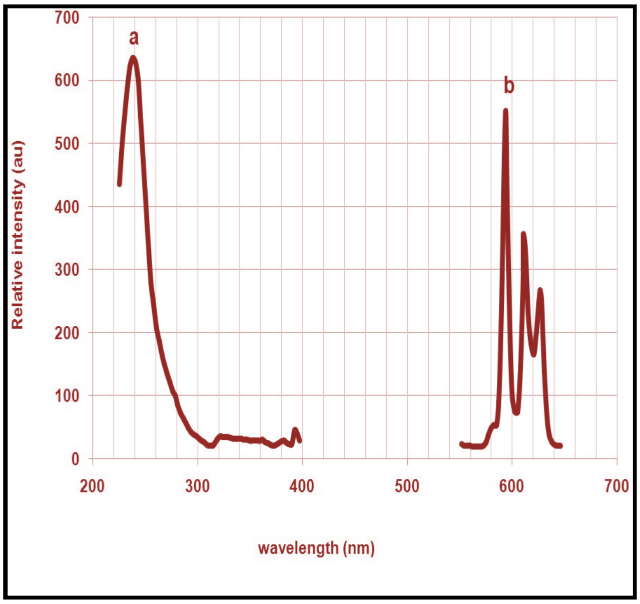

Figure 3. (a) Excitation spectra of YBO3: Eu^{3+} for 595nm emission, (b) Emission spectra of YBO_3: Eu^{3+} for 254 nm excitation

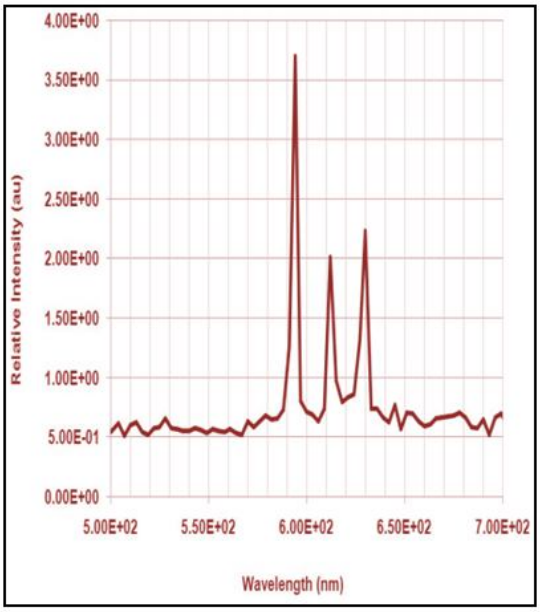

Figure 4. X-ray excited luminescence of YBO_3: Eu^{3+}

RESULT AND CONCLUSION

The photoluminescence properties of the powder samples of $YCaBO_4$:Eu^{3+} and YBO_3:Eu^{3+} has been investigated under UV and VUV excitation. The phosphor shows strong absorption Under UV 254 nm excitation, $YCaBO_4$:Eu^{3+} exhibits intense red emission around 610 nm. Under VUV excitation of 173 nm, the phosphor emits intense red emission around 610 nm which corresponds to the $^5D_0 \rightarrow {}^7F_2$ transition of Eu^{3+} and few weak emissions. These weak emissions could be suppressed by annealing the sample repeatedly at proper temperature, where as YBO_3: Eu^{3+}shows strong absorption in UV and VUV region and exhibits intense orange red emission upon excited by 254 nm UV and 173 nm VUV radiation. The maxima observed at 596 nm and 611 nm in the red region of the spectrum are due to $^5D_0 \rightarrow {}^7F_1$ and $^5D_0 \rightarrow {}^7F_2$ transitions of Eu^{3+} ion. This is in good agreement with the literature value for the VUV excited luminescence could be used for PDP applications.

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