



## Eu ve Tb Katkılandırılmış LuBO<sub>3</sub> ve LaGdPO<sub>4</sub> 'ın Radyolüminesans Özelliklerinin Araştırılması

İsrafil ŞABİKOĞLU\*<sup>1</sup>

<sup>1</sup>Celal Bayar Üniversitesi, Fen Edebiyat Fakültesi, Fizik Bölümü, 45140, Manisa

(Alınış Tarihi: 15.09.2013, Kabul Tarihi: 15.11.2013)

### Anahtar Kelimeler

Radyolüminesans  
LuBO<sub>3</sub>  
LaGdPO<sub>4</sub>  
XRD  
Hidrotermal sentez

**Özet:** Lütisyum ortoborat (LuBO<sub>3</sub>) ve lantan, gadolinyum fosfat (LaGdPO<sub>4</sub>) malzemeleri, borat ve fosfat bileşikleri arasından, sergiledikleri üstün lüminesans özelliklerinden dolayı, optoelektronik cihazlarda kullanılabilecek yeni nesil ev sahibi malzemelerdir. Hidrotermal yöntem ile sentezlenen nano yapılarda, mikro boyuta göre, fiziksel ve kimyasal farklılıklar meydana gelmektedir. Farklı nadir toprak elementleri ile katkılandırılarak yapılan nano yapıların optik özellikleri de değiştirilebilmektedir. Hidrotermal sentez yöntemi, düşük sıcaklık ve yüksek basınç altında, yüksek saflıkta ve nano boyutlarda tek kristal üretimini sağlayan bir sentezleme türüdür. Bu çalışmada, LuBO<sub>3</sub>:NTE ve La,GdPO<sub>4</sub>: NTE (NTE:Eu, Tb) örneklerinin X-ışını kırınımı (XRD) ve radyolüminesans (RL) özellikleri incelenmiştir. Eu ve Tb için tipik elektronik geçişleri gözlenmiştir.

## Investigation of Radioluminescence Properties of Eu and Tb Doped LuBO<sub>3</sub> and LaGdPO<sub>4</sub>

### Keywords

Radioluminescence  
LuBO<sub>3</sub>  
LaGdPO<sub>4</sub>  
XRD  
HydrothermalSynthesis

**Abstract:** In the family of borate and phosphate, orthoborate lutetium (LuBO<sub>3</sub>), and lanthanum gadolinium phosphate (LaGdPO<sub>4</sub>) materials are the new generation materials in optoelectronic devices due to their superior luminescence properties. Some physical and chemical differences occur in nanostructures synthesized by hydrothermal method according to micro-sized structure. Optical properties of nano-sized structures can be changed owing to different doping with rare earth element in to the host material. The hydrothermal synthesis method is a type of synthesis that allowing the production of nano-sized single crystal in high purity under low temperature and high pressure. In this study, radioluminescence and XRD properties of LuBO<sub>3</sub>:REE and La,GdPO<sub>4</sub>: REE (REE: Eu, Tb) materials are investigated. It is observed that typical electronic transition for Eu and Tb.

### 1. Introduction

Today, nanoparticles or nano-sized materials can be produced by various methods, in particularly; they play an important role in the production of optoelectronic and photonic materials. The physical properties of nano-sized material can be changed in quantum mechanical, for example, a semiconductor material can show conductor behavior in the nano-scale [Yang et.al 2008] or their luminescence peaks can shift that blue range [Zhang et.al 1999]. The nano-sized powder with high purity material can be obtained using autoclaves by hydrothermal synthesis method. The metal salts that used in synthesis solve in the liquid form under the high pressure. The nano-sized crystals have been occurred while the solved metal salts are switching between cold zone and hot zone of the autoclave. Nowadays, the orthoborate

phosphors are very often used materials in optoelectronic devices and its application due to their high ultraviolet and optical threshold damage, low synthesis temperature, continuity of characterization [Zhang 1999; Balcerzyk2000]. The orthoborates that doped with variety rare earth element (REE) have good luminescence efficiency. Especially, they can use in mercury-free fluorescent lamp and various projection devices [Li 2006; Kim 2002].

In recent years, it has been promising due to the orthoborate and orthophosphate compositions can be used in scintillator, phosphor, laser, compact fluorescent lamp etc. that have high density, damage threshold and green emitting [Şabikoğlu 2011; Balcerzyk 2000].

\* İlgili yazar: [israfil.sabikoglu@cbu.edu.tr](mailto:israfil.sabikoglu@cbu.edu.tr)

In this study, Europium (Eu) and Terbium (Tb) doped LuBO<sub>3</sub> and La,GdPO<sub>4</sub> phosphors are synthesized using hydrothermal synthesis method. The crystal structure of all material has been investigated and radioluminescence (RL) properties of these materials have been obtained.

## 2. Material Method

Lu<sub>2</sub>O<sub>3</sub>, Tb<sub>4</sub>O<sub>7</sub>, Eu<sub>2</sub>O<sub>3</sub>, Gd<sub>2</sub>O<sub>3</sub>, La<sub>2</sub>O<sub>3</sub> oxides and (NH<sub>4</sub>)<sub>2</sub>HPO<sub>4</sub> (Sigma Aldrich) compositions are used for Eu and Tb doped LuBO<sub>3</sub> and La,GdPO<sub>4</sub> phosphors. The oxides are mixed with HCl or HNO<sub>3</sub> then H<sub>3</sub>BO<sub>3</sub> and deionized water are added into the solution. For control of the pH level, NH<sub>3</sub>·H<sub>2</sub>O solution is added in to the solution drop by drop. The solution is mixed on the magnetic stirrer until gelation occurs. Finally, all solution is transferred to the 50ml autoclave and heated 200°C at 24-32 hours. After the synthesis procedure, all material is washed several times with water and alcohol and filtered. Last procedure, all material is dried in the furnace at 100°C.

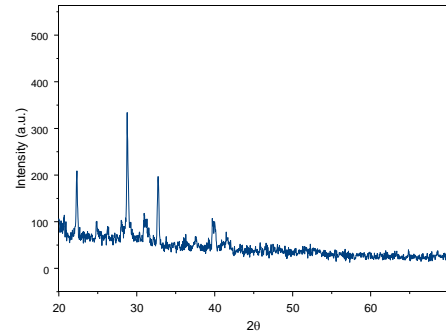
The XRD measurement of the materials are measured on İzmir Institute of Technology at Philips X-pert Pro. The radioluminescence properties of the materials are measured in Celal Bayar University Luminescence Laboratory on room temperature.

## 3. Results and Discussion

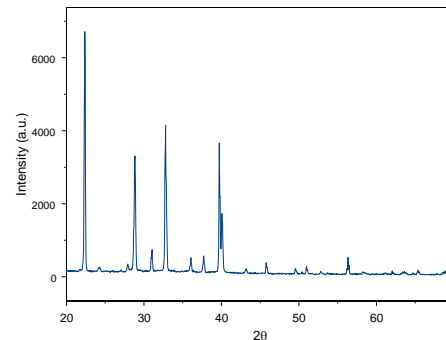
### 3.1. XRD Results

It can see that according to XRD results (Figure 1-4), all material have crystallite shape that made by Hydrothermal synthesis system. La,GdPO<sub>4</sub> composition has complex crystal structure and it is hard to synthesis this composition. According to Figure 1 and 3, there is difference between two La,GdPO<sub>4</sub> materials. It can see that in Figure 3, the La,GdPO<sub>4</sub> material has been occurred, unless the peaks those seen in Figure 1 are very weak in comparison to the other material. The Eu doped La,GdPO<sub>4</sub> phosphor material was not obtained exactly.

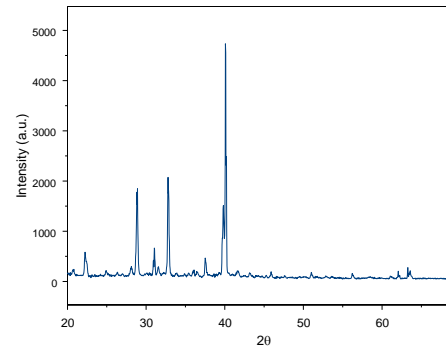
XRD graphs of LuBO<sub>3</sub> are similar each other (Figure 2, 4). As seen in Figure 2, Eu doped LuBO<sub>3</sub> material has been synthesized. However, it can see that there is a difference situation in Figure 4. Some impurities have been occurred in this material because of the last washing and drying procedure may not have been very well.



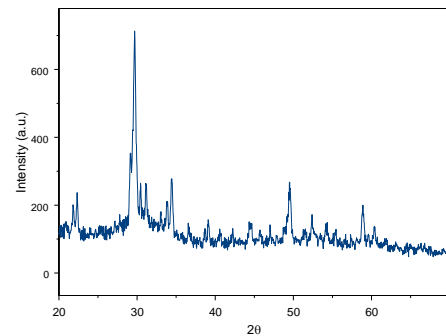
**Figure 1.** The XRD graph of La,GdPO<sub>4</sub>:Eu.



**Figure 2.** The XRD graph of LuBO<sub>3</sub>:Eu.



**Figure 3.** The XRD graph of La,GdPO<sub>4</sub>:Tb.



**Figure 4.** The XRD graph of LuBO<sub>3</sub>:Tb.

### 3.2. RL Results

Eu and Tb lanthanides have most electronic transition [Gruber 2011; Taikar 2012]. In generally, 4f-4f lines are distinctive arising from electronic transition of trivalent lanthanides. The 650nm peak is the most characteristic peak of the LaGdPO<sub>4</sub>:Eu (Figure 5). All peak are overlapped arise form lanthanum (La), gadolinium (Gd) and europium (Eu). Especially, La and Gd have typical electronic transition between at 600-650nm, the Gd has two electronic transition at quadratic 4f-4f at 602nm and 628nm that arise from  ${}^6P_{7/2} \rightarrow {}^8S_{7/2}$  and  ${}^6P_{5/2} \rightarrow {}^8S_{7/2}$  [Wu et.al 2007]. Including the fact that the Eu characteristic electronic transition, there are specific/complex electronic transitions in this range.

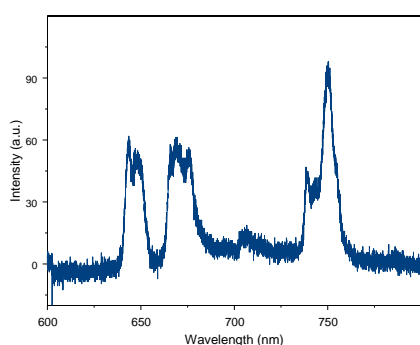


Figure 5. The RL spectrum of La,GdPO<sub>4</sub>:Eu.

The vaterite type material is composed for LuBO<sub>3</sub>:Eu according to Figure 2. The RL spectrum of the LuBO<sub>3</sub>:Eu can be shown in Figure 6. The typical  ${}^5D_0 \rightarrow {}^7F_3$  transition is seen at 650nm and 675nm, also low intensity the  ${}^5D_1 \rightarrow {}^7F_5$  that energy splitting transition is seen at 750nm nearby infrared region [Gruber 2011; Dwivedi 2013].

The RL spectrum of the LuBO<sub>3</sub>:Tb is shown in Figure 7. Four different peaks can be seen at 485, 545, 580 and 625nm. These electronic transitions are allowed and have 4f-4f arise from  ${}^5D_4 \rightarrow {}^7F_6$ ,  ${}^5D_4 \rightarrow {}^7F_5$ ,  ${}^5D_4 \rightarrow {}^7F_4$ , and  ${}^5D_4 \rightarrow {}^7F_3$  respectively.

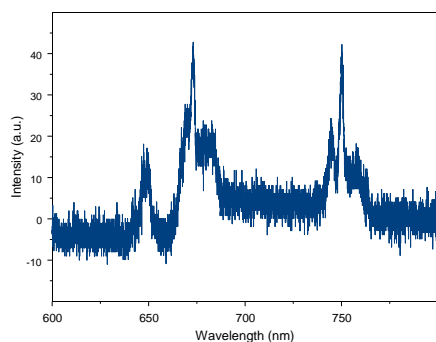


Figure 6. The RL spectrum of LuBO<sub>3</sub>:Eu.

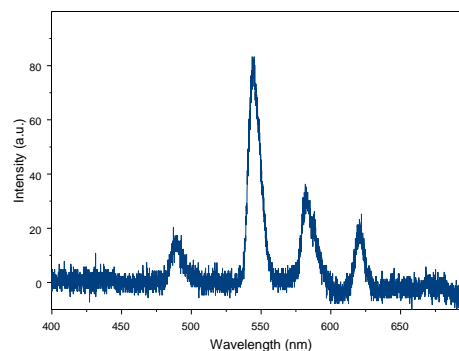


Figure 7. The RL spectrum of LuBO<sub>3</sub>:Tb.

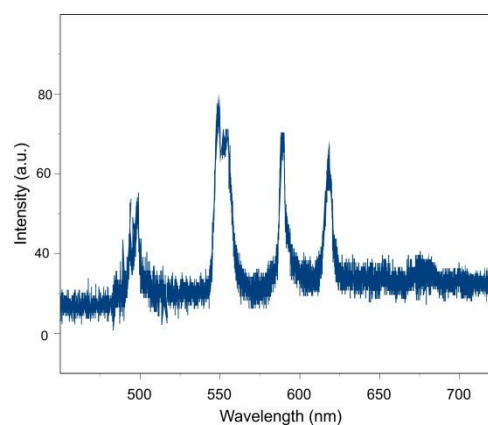


Figure 8. The RL spectrum of La,GdPO<sub>4</sub>:Tb.

As seen in Figure 8, Tb doped LaGdPO<sub>4</sub> material has 4 peaks at ~490, 545, 585 and 620nm. All peaks accommodate with characteristic peaks of Tb at 4f-4f,  ${}^5D_4 \rightarrow {}^7F_j$  [Wu et.al 2007; Wu et.al 2002].

### 4. Conclusion

In this study, Eu and Tb doped La,GdPO<sub>4</sub> and LuBO<sub>3</sub> materials have been synthesized and investigated properties of RL. The XRD results of the some composites are not expected; nevertheless they have glow peaks in visible region. The process of the washing with water and alcohol and drying may have influenced to the XRD results during the synthesizing, so the new procedure of the washing (longer washing) or more filtering ( 4-5 times) can be apply during the synthesizing for the better results of the XRD. As seen in RL figures, all materials have glow peak at visible region. Eu and Tb doped LaGdPO<sub>4</sub> have complex electronic transition because of include another lanthanide Gd. This means, some electronic transition can be occurred from Gd to other lanthanides (Eu, Tb). Likewise, LuBO<sub>3</sub> composites have glow peaks at visible region. Especially, Eu doped LuBO<sub>3</sub> material has energy splitting transition at nearby infrared region.

This study is supported by Celal Bayar University, Coordinator of Scientific Research Projects, and the Project number is 2012-02.

## References

Balcerzyk M, Gontarz Z, Moszynski M, Kapusta M (2000) Future hosts for fast and high light output cerium doped scintillator, *J. Lumin.*, 87–89, 963–966.

Bessière A, Benhamou RA, Wallez G, Lecointre A, Viana B (2012) Site occupancy and mechanisms of thermally stimulated luminescence in Ca<sub>9</sub>Ln(PO<sub>4</sub>)<sub>7</sub>, *Acta Materialia* 60, 6641–6649.

Dwivedi Y and Zilio SC (2013) Infrared cascade and cooperative multicolor upconversion emissions in Y<sub>8</sub>V<sub>2</sub>O<sub>17</sub>:Eu:Yb nanophosphors, *Optic Express* 21-4, 4717.

John B. Gruber et.al (2011) Spectroscopic analysis of Eu<sup>3+</sup> in single-crystal hexagonal phase AlN *J. Applied Physics* 110, 023104.

Kim KN, Jung HK, Park HD (2002) Synthesis and characterization of red phosphor (Y,Gd)BO<sub>3</sub>:Eu by the coprecipitation method, *J. Mater. Res.*, 17, 907.

Li ZH, Zeng JH, Chen C, Li YD (2006) Synthesis and characterization of red phosphor (Y,Gd)BO<sub>3</sub>:Eu by the coprecipitation method, *J. Cryst. Growth*, 286, 487.

Şabikoğlu İ (2011) Katkılandırılmış Lityum Silikat camı seramiklerin sentezlenmesi ve lüminesans özelliklerinin incelenmesi, Doctorate Thesis, Celal Bayar University, Institute of Science 178p.

Taikar DR, Joshi CP, Moharil SV, Muthal PL, Dhopte SM (2012) Synthesis and luminescence of La<sub>2</sub>BaZnO<sub>5</sub> phosphors *J.of Lumin* 11022.

Wu C, Wang Y (2007) Hydrothermal synthesis and luminescent properties of (La,Gd)PO<sub>4</sub>:Tb phosphors under VUV excitation, *Materials Letters* 61, 2416–2418.

Wu X et.al (2002) Vacuum ultraviolet optical properties of (La,Gd)PO<sub>4</sub>:RE<sup>3+</sup> (RE=Eu, Tb), *Mater.Res.Bull.* 37, 1531-1538.

Yang J, Zhang C, Wang L, Hou Z, Huang S, Lian H, Lin J (2008) Hydrothermal synthesis and luminescent properties of LuBO<sub>3</sub>: Tb<sup>3+</sup> microflowers, *J.SolidSt.Che.*, 181, 2672.

Zhang L et.al. (1999) Fast fluorescence and scintillation properties of cerium and praseodymium doped lutetium orthoborates, *Radiat. Eff. Defects Solids*, 150, 47.