

Synthesis and Photoluminescence Study of RE³⁺Co-Doped Y₃Al₅O₁₂ Phosphor

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Abstract—For this study, germanium, europium, terbium co-doped yttrium aluminum garnet (YAG) phosphor powders were prepared via the standard solid state reaction process. The characteristics of the synthesized powders were investigated by means photoluminescence. The photoluminescence spectra of the YAG were investigated to determine the energy level of electron transition related to luminescence processes. There were two peaks in the excited spectrum, and the major one was a broad band of around 260 nm. Also, the YAG showed two emission peaks in the range of 500-600 and 600-nm, respectively, and had maximum intensity at 545 nm.

Keywords-YAG, Solid state reaction, Phosphor, Luminescence, Transition

1. INTRODUCTION

Yttrium aluminum garnet (Y3Al5O12, YAG) is an advanced ceramic with interesting optical and mechanical properties [1]. A host crystal with an yttrium aluminum garnet structure has the advantage of a relatively stable lattice and large thermal conductivity. Thus, YAG can be used as the host lattice for a number of rugged phosphor systems [2]. It is an important crystal for fluorescence and solid state lasers. Eu, Tb-activated YAG phosphor has luminescence characteristic narrow band phosphor suitable for contrast-enhanced display applications in high ambient illumination conditions [3]. Hence, YAG:Tb is one of the promising phosphor candidates that may be used in projection cathode ray tubes, field emission display, and scintillation and electroluminescence applications [4]. To enhance the brightness and resolution of these displays, it is important to develop phosphors with high quantum efficiency, controlled morphology, and small particle sizes. It has been found that the luminescence efficiency of YAG: Tb phosphor depends on the synthesis method, which can lend itself to the formation of single-phase YAG. Commercial phosphors such as YAG:Tb are typically prepared via the conventional solid-state reaction at high temperatures (>1400 °C) [5-8].

2. EXPERIMENTAL

To prepare $Y_3Al_5O_{12}$ with various concentrations of Ge (3.0%), Eu (2.5 mol%) and Tb (3.0 mol%), stoichiometric amounts of reactant mixture is taken in alumina crucible and is fired in air at 1200°C for 2 hour in a muffle furnace. The Tb, Eu³⁺ activated $Y_3Al_5O_{12}$ phosphors was prepared via high temperature modified solid state

diffusion. The starting materials Y_2O_3 , Al_2O_3 , were used to prepare the host phosphor and GeO_2 , Tb_4O_7 , Eu_2O_3 as dopants. The mixture of reagents was grounded together for 45 minute to obtain a homogeneous powder. Powder was transferred to alumina crucible, and then heated in a muffle furnace at 1200 °C for 2 hr. The phosphor materials were cooled to room temperature naturally.

The samples were characterized by using Photoluminescence (PL). The photoluminescence (PL) emission and excitation spectra were recorded at room temperature by use of a Shimadzu RF-5301 PC spectrofluorophotometer

3. RESULTS AND DISCUSSION

3.1 Photoluminescence Study of Ge (3.0%), Eu (2.5%) Co-Doping Y₃Al₅O₁₂ Phosphor

Fig.1 is the PLE & PL spectra of $Y_3Al_5O_{12}$:Ge (3.0%), Eu (2.5%) co-doped phosphor. The PLE spectrum was measured by monitoring ${}^5D_0 \rightarrow {}^7F_J$ emission of Eu³⁺ at 616nm. The PLE spectrum exhibits broad excitation centered at 265nm with high intensity. A series of much weaker intra-4*f*6 electronic transitions of Eu³⁺ in the longer wavelength region as marked in the figure were observed.

The PL spectra shows emission measured from 350 - 650nm range, observed several peaks 368, 398, 420, 431, 451, 468, 474, 483, 525, 592, 596, 600, 609, 616 and 630nm under 254, 265nm excitation. The 368nm peak attributed to crystal field and the 398nm peak is may be due to Ge ion and the remaining peaks are attributed to Eu³⁺ ion transitions from ${}^{5}D_{j} \rightarrow {}^{7}F_{j}$. Among these emissions high intense MD and ED transitions are observed. The MD and ED transitions of Eu³⁺ ion emission under 265nm are high.

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Fig. 1: PLE & PL spectra of Ge (0.1%) & Eu (2.5%) co-doped Y₃Al₅O₁₂ phosphor

3.2 Photoluminescence Study of Tb (3.0%), Eu (2.5%) Co-Doping Y₃Al₅O₁₂ Phosphor

Fig.2 is the PLE & PL spectra of $Y_3Al_5O_{12}$: Tb (3.0%), Eu (2.5%) co-doped phosphor. The PLE spectrum was measured by monitoring ${}^5D_0 \rightarrow {}^7F_J$ emission of Eu³⁺ at 616nm. The PLE spectrum exhibits broad excitation centered at 260nm with high intensity.

The PL spectra shows emission measured from 350 - 650nm range, observed several peaks 368, 398, 420, 431, 451, 468, 474, 483, 525, 592, 596, 600, 609, 616 and 630nm under 254, 265nm excitation. The 368nm peak attributed to crystal field and the 398nm peak is may be due to Ge ion and the remaining peaks are attributed to Eu³⁺ ion transitions from ${}^{5}D_{j} \rightarrow {}^{7}F_{j}$. Among these emissions high intense MD and ED transitions are observed. The MD and ED transitions of Eu³⁺ ion emission under 265nm are high. From the PL spectrum the characteristic emission at 544nm with good intensity is observed due to the ${}^{5}D_{4} \rightarrow {}^{7}F_{5}$ transition.



Fig. 2: PLE & PL spectra of Tb (3.0%) & Eu (2.5%) co-doped Y₃Al₅O₁₂ phosphor



Fig. 3: PLE & PL spectra of Ge (0.1%), Tb (3.0%) & Eu (2.5%) co-doped Y₃Al₅O₁₂ phosphor

3.3 Photoluminescence Study of Ge(3.0%), Tb (3.0%), Eu (2.5%) Tri-Doping Y₃Al₅O₁₂ Phosphor

Fig.3 is the PLE & PL spectra of $Y_3Al_5O_{12}$: Ge(3.0%), Tb (3.0%), Eu (2.5%) co-doped phosphor. The PLE spectrum was measured by monitoring ${}^5D_0 \rightarrow {}^7F_J$ emission of Eu³⁺ at 616nm. The PLE spectrum exhibits broad excitation centered at 255nm with low intensity.

The PL spectra shows emission measured from 350 - 650nm range, observed several peaks 368, 398, 420, 431, 451, 468, 474, 483, 525, 592, 596, 600, 609, 616 and 630nm under 254, 265nm excitation. The 368nm peak attributed to crystal field and the 398nm peak is may be due to Ge ion and the remaining peaks are attributed to Eu³⁺ ion transitions from ${}^5D_j \rightarrow {}^7F_j$. Among these emissions high intense MD and ED transitions are observed. The MD and ED transitions of Eu³⁺ ion emission under 265nm are high. From the PL spectrum the characteristic emission at 544nm with good intensity is observed from Tb³⁺ ion in the host. However the emission inder 400nm is due to the $Y_3Al_5O_{12}$ phosphor

4. CONCLUSIONS

 $Y_3Al_5O_{12}$ phosphors doped with Ge, Eu, Tb, Eu and Ge,Tb. All the three phosphors exhibited good luminescence when excited with 254 and 265 nm under 400nm which is due to host emission. The other emissions are due to the characteristics emissions of Ge, Eu and Tb in $Y_3Al_5O_{12}$ phosphor. It is concluded that these phosphor may be worth further investigations.

REFERENCES

- [1] T. Martin, A. Koch, Journal of Synchrotron Radiation 13 (2006) 180.
- [2] J. Mares, M. Nikl, N. Solovieva, C. D'Ambrosio, F. de Notaristefani, K. Blazek, P.Maly, K. Nejezchleb, P. Fabeni, G. Pazzi, J.T. de Haas, C.W. van Eijk, P. Dorenbos, Nuclear Instruments and Methods in

Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 498 (2003) 312.

- [3] P.K. Sharma, R.K. Dutta, A.C. Pandey, Journal of Nanoparticles Research 14 (2012) 1
- [4] J. Sanghera, W. Kim, G. Villalobos, B. Shaw, C. Baker, J. Frantz, B. Sadowski, I.Aggarwal, Optical Materials 35 (2013) 693.
- [5] W. Liu, D. Zhang, J. Li, Y. Pan, Y. Bo, C. Li, B. Wang, Q. Peng, D. Cui, Z. Xu, Optics and Laser Technology 46 (2013) 139.
- [6] H. Gross, J. Neukum, J. Heber, D. Mateika, T. Xiao, Physical Review B 48 (1993) 9264
- [7] L. Mancic, V. Lojpur, I. Barosso, M.E. Rabanal, O. Milosevic, European Journal of Inorganic Chemistry 2012 (2012) 2716.
- [8] M.L. Keith, R. Roy, American Mineralogist 39 (1959) 1.
- [9] O. Yamaguchi, K. Takeoka, K. Hirota, H. Takano, A. Hayashida, Journal of Material Science 27 (1992) 1261
- [10] B. Basavalingu, M.S. Vijaya Kumar, H.N. Girish, S. Yoda, Journal of Alloys and Compounds 552 (2013) 382
- [11] T.B. de Queiroz, C.R. Ferrari, D. Ulbrich, R. Doyle, A.S.S. de Camargo, Optical Materials 32 (2010) 1480
- [12] N. Kalivas, I. et al Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 569 (2006) 210.
- [13] H. Gao, Y. Wang, Materials Research Bulletin 42 (2007) 921.
- [14] G. Kim, H. Park, T. Kim, Materials Research Bulletin 36 (2001) 1603.

- [15] Z. Liu, J. Ma, Y. Sun, Z. Song, J. Fang, Y. Liu, C. Gao, J. Zhao, Ceramics International 36 (2010) 2003.
- [16] D Tawde, M Srinivas, KVR Murthy, physica status solidi (a) 208 (4), 803-807
- [17] KVR Murthy, L Rey, P Belon Journal of luminescence 122, 279-283
- [18] BS Chakrabarty, KVR Murthy, TR Joshi Turkish Journal of Physics 26, 193-198
- [19] KVR Murthy, Recent Research in Science and Technology 4 (8), 2013
- [20] P Page, KVR Murthy, Philosophical Magazine Letters 90 (9), 653-662
- [21] MR Rao, BS Rao, NP Rao, K Somaiah, KVR Murthy, Indian Journal of Pure & Applied Physics 47 (6), 456
- [22] KVR Murthy, YS Patel, AS Sai Prasad, V Natarajan, AG Page, Radiation measurements 36 (1), 483-485
- [23] KVR Murthy, JN Reddy, Thermoluminescence basic theory application and experiment, Feb. 2008, Pub:Nucleonix, Hyderabad
- [24] MC Parmar, WD Zhuang, KVR Murthy, XW Huang, YS Hu, V Natarajan, Indian Journal of Engineering and Materials Sciences 16 (3), 185
- [25] MD Sastry, M Gaonkar, S Mane, S Athavale, KVR Murthy, S Desai, Diamond and Related Materials 17 (7), 1288-1291
- [26] V Dubey, J Kaur, S Agrawal, NS Suryanarayana, KVR Murthy, Superlattices and Microstructures 67, 156-171
- [27] G.L.Sudharani, K.Suresh and K.V.R.Murthy, International Journal of Luminescence and its Applications, ISSN: 81-6717-806-5 (2014)