



LUMINESCENCE BEHAVIOUR OF Dy^{3+} ION DOPED MAGNESIUM SULFOBORATE PHOSPHOR FOR WHITE LIGHT EMITTING DIODES

*¹Dalhatu, A. S., ²Hussin, R., ³Ibrahim, B., ²Yamusa, Y. A. and ¹Baballe, A.

*¹Department of Physics, Bauchi State University Nigeria, 65 Gadau, Bauchi, Nigeria

Phone: +2348069364724: Email: sadgambaki@yahoo.com

Phone: +2348139099865: Email: ababalle@yahoo.com

²Department of Physics, Faculty of Science, Universiti Teknologi Malaysia, 81310 Skudai, Johore Bahru, Johore, Malaysia

Phone: +60199898425: Email: roslihussin@utm.my

Phone: +601136983790: Email: yamusaabdullahi@yahoo.com

³Department of Physics, School of Sciences, Kaduna State College of Education Gidan waya, Kafanchan, Nigeria

Phone: 07068102222: Email: ibshekwolo@yahoo.com

ABSTRACT

Several studies showed the interesting properties of trivalent lanthanide ions when doped in various types of phosphor. Magnesium sulfoborate phosphor doped with different concentrations of Dy^{3+} were synthesized using solid-state reaction method at 850 °C for 4 hours. The samples were characterized by X-ray Diffraction (XRD). The excitation and luminescence properties of $MgO-SO_4-B_2O_3:Dy^{3+}$ were determined. The emission spectrum of Dy^{3+} ion doped $MgO-SO_4-B_2O_3$ phosphor exhibit three bands at 480 nm, 573 nm and 660 nm with excitation of 386 nm due to $^4F_{9/2} \rightarrow ^6H_{15/2}$, $^6H_{13/2}$ and $^6H_{11/2}$ of Dy^{3+} transitions, respectively. The excitation spectrum of Dy^{3+} ion doped $MgO-SO_4-B_2O_3$ phosphor display several bands at 347 nm, 362 nm, 386 nm, 426 nm, 449 nm and 469 nm with emission of 573 nm, which is in agreement with the ultraviolet LED (349.9–410 nm) and blue LED (450–470 nm). An intense in the emission peak at 573 nm in the yellow region was observed with the 0.5 Dy_2O_3 . The luminescence properties of phosphor show that $MgO-SO_4-B_2O_3:Dy^{3+}$ phosphor could be potentially used as white LEDs.

Keywords: Magnesium Sulfoborate, Phosphors, White LEDs, Luminescence properties

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INTRODUCTION

White light emitting diodes (WLEDs) in the field of solid-state lighting are promising new emitting light source due to their long life time, environmental-friendliness and energy saving devices (Krames *et al.*, 2007; Pavani *et al.*, 2011). The previous study used three methods in order to generate white light, one by the use of primary tricolor phosphor, two by UV-LED+RGB and third by blue LED combined with yellow emitting phosphor as shown in Figure 1 (Ratnam *et al.*, 2009; Ye *et al.*, 2010). White light emission resulted from a single-phase phosphor with high luminous efficiency is required (Ratnam *et al.*, 2010). Therefore, single phase of white-emitting luminescence is UV required for -pumped white LEDs to enhance the luminescence and efficiency (Nagpure *et al.*, 2010). Phosphors doped rare earth ions have been studied by many researchers due to white light emission application. Dysprosium ions have two intense bands in the blue (484 nm) from transition of ${}^4F_{9/2} \rightarrow {}^6H_{15/2}$ and yellow (575 nm) from transition of ${}^4F_{9/2} \rightarrow {}^6H_{13/2}$ (Zhang *et al.*, 2012). There are many reports on hosts doped with trivalent dysprosium ions, such as, phosphate $\text{NaCaPO}_4:\text{Dy}^{3+}$ (Ratnam *et al.*, 2010), borates $\text{Ba}_2\text{LiB}_5\text{O}_{10}:\text{Dy}^{3+}$ (Liu *et al.*, 2011), silicate $\text{CaMgSi}_2\text{O}_6:\text{Dy}^{3+}$ (Chen *et al.*, 2009) and some oxide $\text{Y}_2\text{O}_3:\text{Dy}^{3+}$ (Jayasimhadri *et al.*, 2010). Among these phosphors, borate phosphors have been attracted much attention because of its a simple preparation, stable crystal structure, high thermal stability and cheap raw material (Li *et al.*, 2010). However, to the best of our knowledge, there has been no report on $\text{MgOSO}_4\text{B}_2\text{O}_3:\text{Dy}^{3+}$ for application in white LEDs. Therefore, in this paper, a novel white $\text{MgOSO}_4\text{B}_2\text{O}_3:\text{Dy}^{3+}$ phosphor was prepared by conventional solid-state reaction method and their photoluminescence properties, such as excitation and emission were presented.

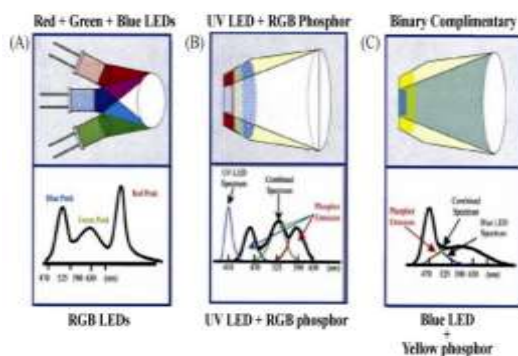


Figure 1: Diagram showing three way to generate white light from LEDs: (A) red + green + blue-LEDs, (B) UV-LED + RGB phosphors, and (C) blue-LED + yellow phosphor (Ye *et al.*, 2010)

EXPERIMENTAL PROCEDURE

A series of $\text{MgO-SO}_4\text{-B}_2\text{O}_3:\text{Dy}^{3+}$ samples were synthesized using solid state reaction method. Analytical reagent MgO , H_2SO_4 , B_2O_3 and Dy_2O_3 (99.99%) were used as the raw materials. The Dy^{3+} ion concentrations ranged from 0.1 mol% to 0.7 mol% in $\text{MgO-SO}_4\text{-B}_2\text{O}_3$. After raw materials were mixed and grained thoroughly in an agate mortar, the mixed powders were placed into alumina crucible and heat at $800\text{ }^\circ\text{C}$ for 2 hours inside the electric furnace. The samples were cooled to room temperature, thus the $\text{MgO-SO}_4\text{-B}_2\text{O}_3:\text{Dy}^{3+}$ phosphors were obtained. The phosphor samples are grained to powder form for characterization. The structure of samples of Dy^{3+} ion doped $\text{MgO-SO}_4\text{-B}_2\text{O}_3$ phosphor were characterized by powder X-ray diffraction (XRD) (Rigaku D/MAX-2500, Cu $\text{K}\alpha$, 40 kV, 150 mA). The emission

as well as excitation were measured using SHIMADZU RF-540 UV spectrophotometer. The luminescence characteristics of these phosphors were measured at room temperature.

RESULTS AND DISCUSSION

XRD, EDX AND SEM ANALYSIS

MgO-SO₄-B₂O₃ has cubic structure with F-43c (219) space group, and its lattice parameters are a=12.0970 nm, b=12.0970 nm, c=12.0970 nm (PDF card No: 00-026-1254). The XRD pattern of the MgO-SO₄-B₂O₃ with concentration ranged from 0.1 to 0.7 mol% of Dy³⁺ as shown in Figure 2. We observed that all samples have the same pattern, and index to the MgOSO₄B₂O₃ crystalline phase. The results here also show that Dy³⁺ ions do not form any new phases in the synthesis process.

Figure 3 shows the energy dispersive X-ray (EDX) of the MgO-SO₄-B₂O₃: 0.5Dy³⁺ phosphor. In EDX the phosphor consists of Mg, S, O, B and Dy elements, thus the EDX spectrum shows all the major elements used in the prepared sample.

Figure 4 shows the scanning electron microscope (SEM) image of the sample. In SEM the shape of the phosphor particle of the 0.5% Dy₂O₃ indicates the crystalline nature of the sample, and homogeneity is observed.

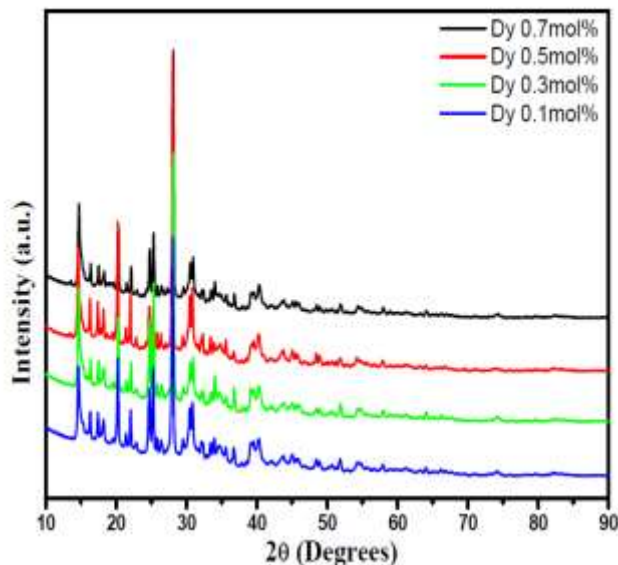


Figure 2: XRD pattern of MgO-SO₄-B₂O₃:Dy³⁺

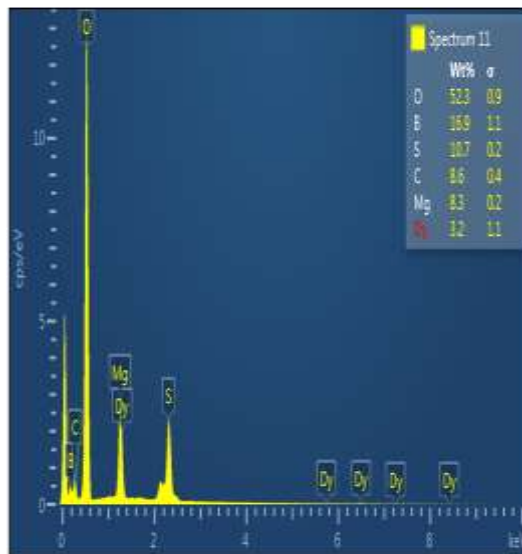


Figure 3: EDX spectra of MgO-SO₄-B₂O₃: 0.5 Dy³⁺phosphor

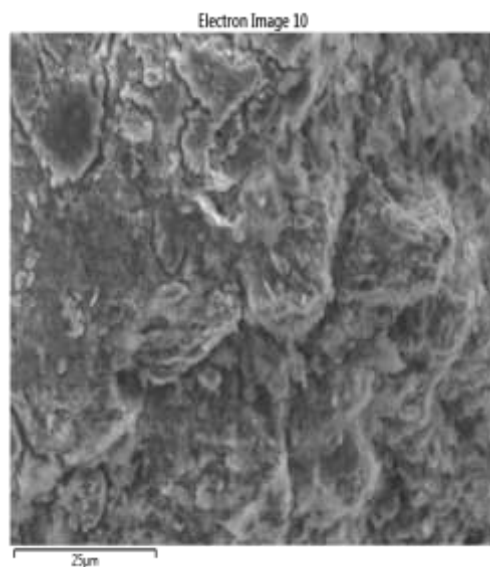


Figure 4: SEM image MgO-SO₄-B₂O₃: 0.5 Dy³⁺phosphor

LUMINESCENCE PROPERTIES

The excitation spectra of the MgO-SO₄-B₂O₃:Dy³⁺ phosphor is shown in Figure 5. The excitation spectrum for 573 nm emission exhibit six bands at 347 nm, 362 nm, 386 nm, 426 nm, 449 nm and 469 nm (Li *et al.*, 2008). The emission spectra of MgO-SO₄-B₂O₃:Dy³⁺ phosphor shows three bands at 480 nm, 573 nm and 660 nm under excitation 386 nm, which correspond to the transitions of ⁴F_{9/2} → ⁶H_{15/2}, ⁶H_{13/2} and ⁶H_{11/2} of Dy³⁺, respectively, as shown in Figure 6. The emission intensities is increases with increasing Dy³⁺ concentration, and reach the maximum value at 0.5 mol% Dy³⁺, then decrease because of the concentration quenching were observed (Zhang *et al.*, 2013). The excitation and emission spectra show that the white LED can be presented through combining ultraviolet chip with MgO-SO₄-B₂O₃:Dy³⁺ phosphor. Moreover, the phosphor is very suitable for a color converter of white LED that uses blue LED as the

primary light source, i.e., it can be used as a white phosphor excited by UVLED chip to fabricate white light, or by blue LED chip to generate white light.

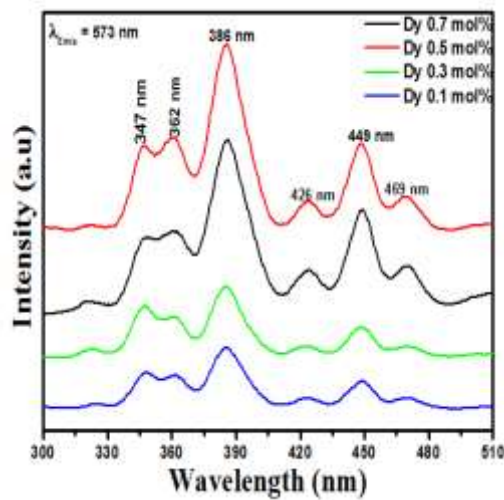


Figure 5: The excitation of MgO-SO₄-B₂O₃:Dy³⁺

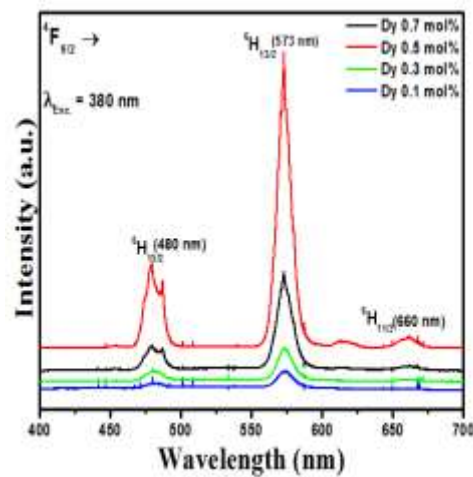


Figure 6: The emission of MgO-SO₄-B₂O₃:Dy³⁺

CONCLUSIONS

Magnesium sulfoborate doped dysprosium ions (MgO-SO₄-B₂O₃:Dy³⁺) has been synthesized using solid state reaction method. XRD analysis shows that the samples show two phases, such as monoclinic and triclinic phase. The emission spectrum of Dy³⁺ ion doped MgO-SO₄-B₂O₃ phosphor exhibit three bands at 480 nm, 573 nm and 660 nm with excitation 386 nm due to ⁴F_{9/2} → ⁶H_{15/2}, ⁶H_{13/2} and ⁶H_{11/2} transitions of Dy³⁺, respectively. The excitation spectrum with the emission of 573 nm have several bands centred at 347 nm, 362 nm, 386 nm, 426 nm, 449 nm and 469 nm, which is in agreement with the ultraviolet LED (350–410 nm) and blue LED (450–470 nm). The optimal luminescence intensity of the MgO-SO₄-B₂O₃:Dy³⁺ phosphor is found at 0.5 concentration of Dy³⁺. The results show that MgO-SO₄-B₂O₃:Dy³⁺ phosphor could be potentially used as white LEDs.

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