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Optical properties of Dy^{3+} activated $MgAl_2B_2O_7$ phosphor using slow evaporation method

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ABSTRACT

Present work reports, synthesis of Dy^{3+} activated $MgAl_2B_2O_7$ (MAB) phosphor by using slow evaporation method. The prepared phosphors have been characterized by XRD, photoluminescence (PL) techniques. The photoluminescence emission spectra of the phosphors having an excitation at around 351 nm (mercury free) showed two distinguishing bands centered at around 484 nm (blue) and 577 nm (yellow) corresponding to $^4F_{9/2}$-$^6H_{15/2}$ and $^4F_{9/2}$-$^6H_{13/2}$ transitions of Dy^{3+}, respectively. These phosphors have strong absorption in the near UV region. Furthermore, the chromaticity coordinate values were estimated from emission spectra of MAB. Moreover, the luminescence quenching noticed at optimum Dy^{3+} concentrations was due to the exchange interaction among the excited Dy^{3+} ions. The observed broad blue and yellow bands in Dy^{3+} ions show that phosphor may be applicable for white-LEDs.

Keywords: Alumino-Borate, rare earth ions, MAB:Dy^{3+}, quenching mechanism.

I. INTRODUCTION

As far as the environmental issues are concern, the requirement for mercury-free fluorescent lamps for general lighting has become fundamental subject for light source manufacturers and researchers. Much attempt has gone into developing mercury-free light sources. The demand has quickly increased and become important to avoid using poisonous ingredients and environmentally unsafe materials [1]. Lamp industries only aspire to reduce content of mercury from fluorescent lamps by using the development of alternative mercury free excitation sources such as plasma devices, light emitting diode devices for general lighting purposes [2-3].

The materials can find potential applications in different aspects as optical amplifiers or lasers, phosphors and photocatalysts, especially when the size of the particles is reduced to nanoscale [4-6] by introducing rare earth (RE) ions into different lattices. Dy3+, as one of the most investigated ions, has been studied extensively because it provides two typical emission bands in blue (480 nm) and yellow (570 nm) regions, which are necessary for full color displays [7-8]. In addition, the emission probability of electric dipole transition is greatly affected by the crystal field and radial integral of 4f and 5d electrons [9]. Thus, it is interesting to study the luminescence properties of Dy^{3+} in different host lattices.

In general, Dy^{3+} exhibits three visible emission bands including the blue emission at 480 nm corresponding to the $^4F_{9/2}\rightarrow{}^6H_{15/2}$ transition, the yellow emission at 575 nm corresponding to the hypersensitive transition $^4F_{9/2}\rightarrow{}^6H_{13/2}$ transition and the feeble red emission at 665 nm corresponding to the transition $^4F_{9/2}\rightarrow{}^6H_{11/2}$ [10-11]. The hypersensitive transition of Dy^{3+} ion is easily influenced by microscopic environment.

The present work reports synthesis, structures, and spectroscopic investigations of $MgAl_2B_2O_7$ phosphor doped with different concentrations of Dy^{3+} ions. To the best of our knowledge, no report exists on luminescent properties of rare earth doped magnesium dialumino-borate phosphor.

II. EXPERIMENTAL

Materials and Method

The powder samples of Dy^{3+} doped MAB were prepared by a solution combustion technique which is described earlier [12-18]. We have successfully synthesized many borate host phosphors using solution combustion synthesis technique. The stoichiometric amounts of high purity starting materials, $Mg(NO_3)_2$ (A.R.), $Al(NO_3)_3.9H_2O$ (A.R.), $Dy(NO_3)_3$ (high purity 99.9%), H_3BO_3 (A.R.), $CO(NH_2)_2$ (A.R.) have been used for phosphor preparation. The stoichiometric amounts of the ingredients were thoroughly mixed in an Agate Mortar, adding little amount of double distilled water to obtain an aqueous homogeneous solution. The aqueous solution was then transferred into a china basin and slowly heated at lower temperature of 70°C in order to remove the excess water. The solution was then introduced into a preheated muffle furnace maintained at 550 °C. The solution boils foams and ignites to burn with flame; a voluminous, foamy powder was obtained. The entire combustion process was over in about 5 min. Following the combustion, the resulting fine powders were annealed in a slightly reducing atmosphere at temperature 750°C for about 90 min. and suddenly cooled to room temperature. The structure of the prepared material was characterized by powder XRD. Powder X-ray diffraction measurements were taken on Rigaku Miniflex II X-ray Diffractometer and compared with the ICDD files. PL & PLE measurements at room temperature were performed on Hitachi F-7000 Spectroflurometer with spectral resolution of 2.5 nm.

III. RESULTS AND DISCUSSION

3.1 XRD pattern of $MgAl_2B_2O_7$ phosphor

In present work, Polycrystalline sample of $MgAl_2B_2O_7$ phosphor was obtained as described above in experimental section. The phase purities of the sample were checked by powder X-ray diffraction. **Fig. 1** shows the XRD pattern of MAB:Dy^{3+} phosphor. The XRD pattern of the as prepared phosphor powder shows good agreement with standard ICDD File. Depending on the ionic radii, for Dy^{3+} (0.912˚A) which is too small to substitute for Mg (0.72 Å) and Al (0.535 Å), thus Dy^{3+} ions may enter into the host lattice to substitute Al^{3+} or Mg^{2+} or locate on the surface of the crystal. Therefore, it would have greater tendency to occupy Aluminum as compared to magnesium sites.

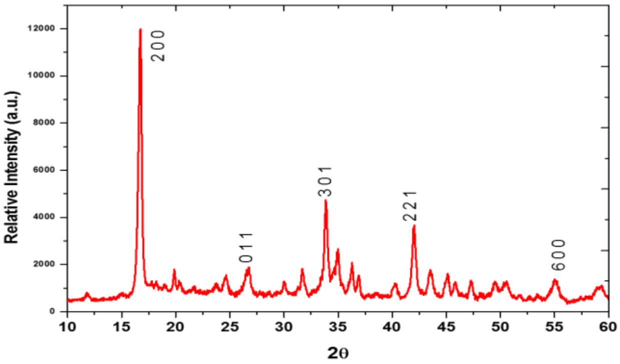

Figure 1. X-ray powder diffraction pattern of $MgAl_2B_2O_7$ host phosphor.

3.2 Photoluminescence investigations

3.2.1. Excitation and emission spectra of Dy^{3+} activated $MgAl_2B_2O_7$phosphor

The luminescence behavior of Dy^{3+} in MAB was examined. Emission spectra were recorded under excitation by 349 nm ($^4K_{17/2}$ state). Two relative intense bands at 484 and 576 nm were observed independently from the excitation wavelengths. They corresponded to $^4F_{9/2}$-$^6H_{15/2}$ (blue), $^4F_{9/2}$-$^6H_{13/2}$ (yellow) transitions of Dy^{3+} ions, respectively. Owing to small energy gaps between all states, the $^4F_{9/2}$ state was efficiently populated by non-radiative relaxation. Then, quite strong yellow and blue emission originating from the $^4F_{9/2}$ state was observed. This phenomenon was due to a large separation (~6000 cm^{-1}) between the $^4F_{9/2}$ state and the next lower lying $^6F_{1/2}$ state and the relative high phonon energy of the host (~1300 cm^{-1}). The intensity of electric dipole transitions strongly depends on the site symmetry in a host crystal. Magnetic dipole transitions are not affected much by the site symmetry because they are parity-allowed [19]. Like Eu^{3+} ions [20], in MAB host, if Dy^{3+} occupies an inversion symmetry site in the crystal lattice, the magnetic dipole transition is the dominant transition. On the contrary, the electric dipole transition will be the dominant transition if Dy^{3+} occupies an unsymmetrical site in the crystal lattice. The $^4F_{9/2}$-$^6H_{13/2}$ transition of Dy^{3+} belongs to electric dipole transitions and $^4F_{9/2}$-$^6H_{15/2}$ transition belongs to magnetic dipole transitions. It can be seen from Fig. 2 that the strong emission line at 484 nm ($^4F_{9/2}$-$^6H_{15/2}$) is the dominant band, which indicates that Dy^{3+} ions occupy the inversion symmetry site.

3.3 Color co-ordinates

The Commission International de'Eclairage (CIE) chromaticity coordination points of the $Mg_{(0.97)}Al_2B_2O_7:_{(0.03)}Dy^{3+}$ phosphor monitored at 349 nm is as shown in Fig. 3. The color chromaticity coordinates are (x=0.292, y=0.323) which is very close to the standard white light NTSC color coordinates (x=0.310, y=0.316).

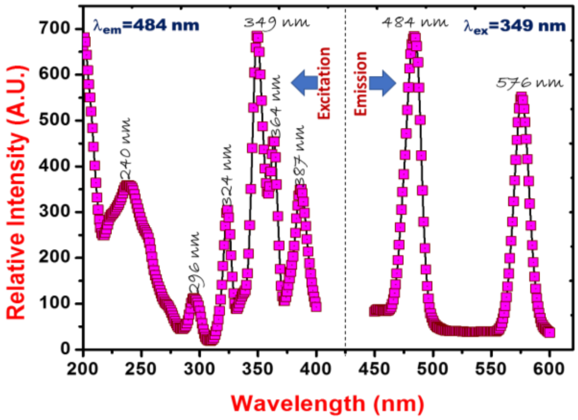

Figure 2. Excitation and emission spectra of $Mg_{(0.97)}Al_2B_2O_7:_{(0.03)}Dy^{3+}$ phosphor.

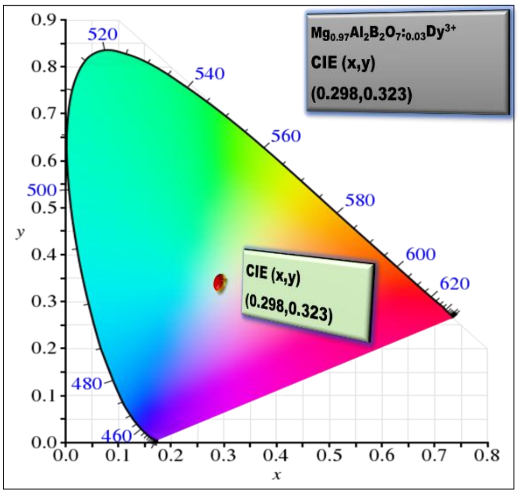

Figure 3. Chromaticity coordinates of $Mg_{(0.97)}Al_2B_2O_7:_{(0.03)}Dy^{3+}$ phosphor in the CIE 1931 chromaticity diagram.

IV. CONCLUSIONS

In conclusions, Dy^{3+} activated MAB phosphor was synthesized by a simple, time saving, low temperature and inexpensive solution combustion technique. It shows an efficient intense absorption band around 349 nm, which match well with the n-UV light-emitting diodes, and shows an emission bands in the blue and yellow regions of the spectrum and it may be helpful for the production of white LEDs. With respect color purity issue, this phosphor has CIE coordinates (x=0.292, y=0.323). The results indicate that MAB:Dy^{3+} is a promising yellowish-blue phosphor for the application in blue LED chips based white-light LEDs.

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VI. REFERENCES

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