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WET CHEMICAL SYNTHESIS OF Tb^{3+} AND Eu^{3+} DOPED $Na_2Sr_2Mg(BO_3)_2F_2$ PHOSPHOR FOR TL DOSIMETER

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Abstract- This paper reports the luminescent properties of Tb^{3+} and Eu^{3+} doped triple mixed borate ($Na_2Sr_2Mg(BO_3)_2F_2$) synthesized by simple wet chemical route. Phosphor has been studied for photoluminescence and thermoluminescence characterizations. PL emission spectra of $Na_2Sr_2Mg(BO_3)_2F_2$:Tb^{3+} shows four peaks showing optimum intensity at 545nm monitored at excitation wavelength 370 nm. The emission spectra exhibit four main bands with the maxima at about 490, 545 (the highest one), 587, and 626 nm, which are due to the transitions from the excited state $^5D_4$ to the ground states ^7F_J (J=6, 5, 4, 3) of Tb^{3+} in the host lattice. PL emission spectra of Eu doped Na_2Sr_2Mg $(BO_3)_2F_2$ phosphor shows two well resolved peaks around 593nm and 618nm which are assigned to $^5D_0 \rightarrow {}^7F_1$ and $^5D_0 \rightarrow {}^7F_2$ transition of Eu^{3+} ion and a peak around 425nm due to $^4f_6{}^5d_1 \rightarrow {}^4f_7$ transition of Eu^{2+} in the blue region of visible spectrum. Thermoluminescence glow curves of $Na_2Sr_2Mg(BO_3)_2F_2$ phosphor doped with Eu^{3+} and Tb^{3+} ions exposed to 0.5 kGy irradiation dose of ^{60}Co are illustrated. TL glow curves consist of only one peak which is fairly symmetrical. Glow peak is observed at 221ºC for Eu^{3+} doped host and TL intensity is about 2 times less than standard $CaSO_4$:Dy. These properties about the phosphor may prove this a good candidate for TL dosimetry.

Keywords- Photoluminescence, thermoluminescence, Glow peak, Dosimetry

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Introduction

In many of the applications of luminescent materials, inorganic solids doped with rare earth impurities are used. To understand how rare earth impurities make various applications possible, it is necessary to know the luminescent characteristics of these materials. Basically there are four important parameters viz. excitation type and spectrum, relaxation to emitting state and the decay time, emission intensity and the emission spectrum, which determine the utility of rare earth doped phosphors. All these parameters may further depend on the concentration and temperature. This dependence is equally important in the context of utility of the phosphors in various applications. One of the striking applications of these is radiation dosimeter.

A radiation dosimeter is a device, instrument or system that measures or evaluates, either directly or indirectly, the quantities exposure, kerma, absorbed dose or equivalent dose, or their time derivatives (rates), or related quantities of ionizing radiation. A dosimeter along with its reader is referred to as a dosimetry system. Thermoluminescence dosimeters are gaining more and more attention due to their several advantages over others.Thermoluminescence (TL) is a well known technique that widely used in the dose measurement of ionizing radiations such as UV, X-rays, gamma rays and ion beam. An ideal TL dosimeter phosphor is expected to possess certain features such as: a relatively simple glow curve having ideally a single peak with its temperature over 200ºC, same TL response for all energies of ionizing radiation, high sensitivity that includes both a high efficiency light emission and a low threshold dose, low fading, good linearity of the TL signal in the specific useful range of radiation dose. Many phosphors have been prepared, in this context recently, nanosize TL materials have also been reported by Numan Salah et.al [1] and Sahare et.al [2].

In this paper we are reporting photoluminescent and Thermoluminescent properties of Tb^{3+} and Eu^{3+} doped $Na_2Sr_2Mg(BO_3)_2F_2$ Phosphor.

Experimental

Pure and rare earth doped $Na_2Sr_2Mg(BO_3)_2F_2$ Phosphor was prepared by a simple wet chemical route.The starting materials used were, $Sr(NO_3)_2$(Merck), NaF(Himedia), $Mg(NO_3)_26H_2O$ (Merck), H_3BO_3 (Merck) Eu_2O_3 andTb_2O_3 (SD Fine). The starting materials were dissolved in double distilled water in separate beakers and stirred vigorously till constituents dissolved completely. The individual solutions were added dropwise and turnwise to ensure homogeneity of solution followed by vigorous stirring. The solution was put on stirrer at 80°C till evaporation of volatile constituents. In this way white crystalline powder of $Na_2Sr_2Mg(BO_3)_2F_2$ phosphor is prepared.

Results and Discussion

XRD Pattern

Figure 1. shows X-ray diffraction pattern of $Na_2Sr_2Mg(BO_3)_2F_2$. As no JCPDS file of this compound is available therefore it might be considered that our compound has acquired the same phase as it shows. The XRD pattern did not indicate the presence of starting constituents and other probable phases which in turn is an indirect evidence for the complete formation of the required compound.

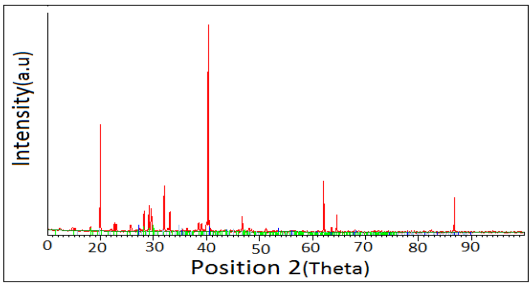

Fig. 1- X-ray powder diffraction pattern of $Na_2Sr_2Mg(BO_3)_2F_2$ host

PL characteristics

The photoluminescence spectra of rare earth(Tb,Eu) doped $Na_2Sr_2Mg(BO_3)_2F_2$ phosphor is (recorded using SHIMADZU RF5301 spectrofluorometer with slit width 1.5 nm) given in figure below. The luminescence behavior of rare earth ions (Dy^{3+}, Sm^{3+}, Eu^{3+}) in magnesium fluoroborate $Mg_3BO_3F_3$ phosphor were first studied by Liu et al. [2], later optical characterization of $Mg_3BO_3F_3$ activated with impurities Dy^{3+} , Gd^{3+} , Eu^{3+} and Pr^{3+} phosphors was done by Van der Voort and Blasse (1991) [3] reported in detail.

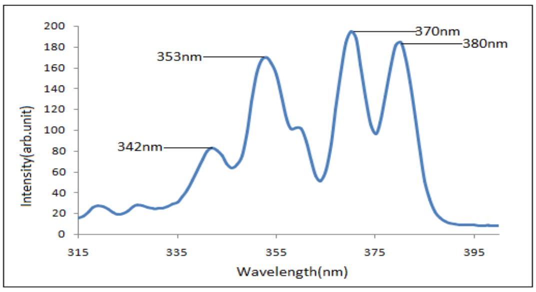

Fig. 2- Excitation spectrum of $Na_2Sr_2Mg(BO_3)_2F_2:Tb^{3+}$

Emission spectra of $Na_2Sr_2Mg(BO_3)_2F_2:Tb^{3+}$ shows four peaks showing optimum intensity at 545nm monitored at excitation wavelength 370nm.

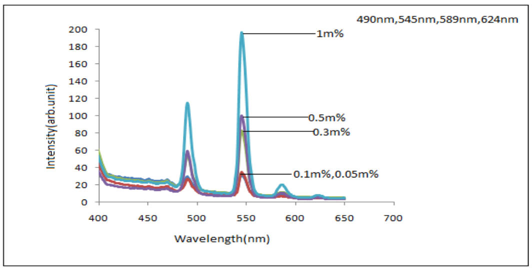

Fig. 3- Emission spectra of $Na_2Sr_2Mg(BO_3)_2F_2:Tb^{3+}$+phosphor monitored at 370nm excitation wavelength

The emission spectra exhibit four main bands with the maxima at about 490, 545 (the highest one), 587, and 626 nm, which are due to the transitions from the excited state $^5D_4$ to the ground states ^7F_J (J=6, 5, 4, 3) of Tb^{3+} in the host lattice. From PL characteristics it can be seen that PL intensity is same for concentrations 0.05m% and 0.1m% and then increase as concentrations are increased further(0.3m%---1m%) shown in fig 5.

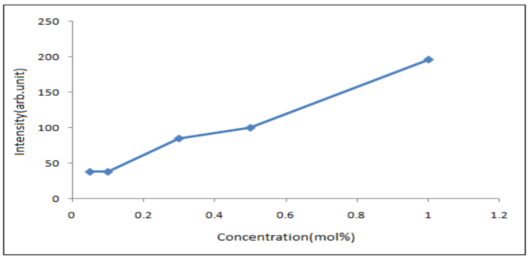

Fig. 4- Concentration of Tb^{3+} ions vs PL peak intensity graph of $Na_2Sr_2Mg(BO_3)_2F_2:Tb^{3+}$ phosphor

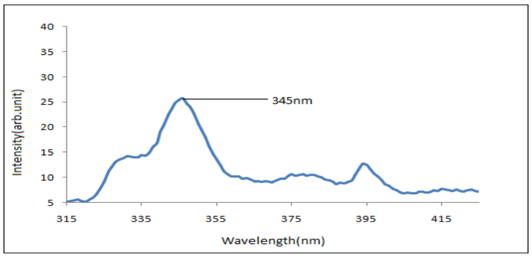

Fig. 5- Excitation spectrum of $Na_2Sr_2Mg(BO_3)_2F_2:Eu^{3+}$

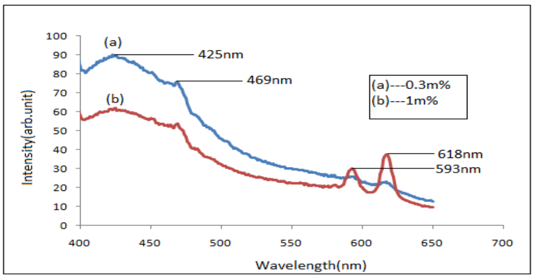

Fig. 6- Emission spectra of $Na_2Sr_2Mg(BO_3)_2F_2:Eu^{3+}$

Figure 5 and 6 depict excitation and emission spectra of $Na_2Sr_2Mg(BO_3)_2F_2:Eu^{3+}$ phosphor with different concentrations under the excitation 345nm wavelength. Two well resolved peaks are observed around 593nm and 618nm,which are assigned to to $^5D_0 \rightarrow ^7F_1$ and $^5D_0 \rightarrow ^7F_2$ transition of Eu^{3+} ion and a peak around 425nm due to $^4f_6{}^5d_1 \rightarrow ^4f_7$ transition of Eu^{2+} in the blue region of visible spectrum.

TL characteristics

Thermoluminescence characterizations are performed on Thermoluminescence Reader Type TL1009 designed and offered by NUCLEONIX SYSTEMS which is a versatile controller based unit, facilitating the user to subject the TL sample under study to the desired heating profile, to record the digitized TL glow curve. This unit stores both integral value and digitized glow curve into EEPROM memory.

The Z_{eff} of $Na_2Sr_2Mg(BO_3)_2F_2$ phosphor is estimated 28.39 and it is very high Z_{eff} value as comparative $CaSO_4$:Dy and LiF:Mg,Cu,P well known standard TLD phosphors. Thermoluminescence glow curves of $Na_2Sr_2Mg(BO_3)_2F_2$ phosphor doped with Eu^{3+} ions and exposed to a 0.5 kGy irradiation dose of ^{60}Co relative to $CaSO_4$:Dy shown in figure 7.TL glow curves consist of only one peak which is fairly symmetrical. the attachment of fluorides due to their large bandgap, are suitable for impurities to create defect centers [4]. There is a prominent glow peak observed at high temperature around 221ºC due to high energy traps and it is very useful for thermoluminescence dosimetry (TLD) phosphor characteristics. The glow curve is similar for all four concentrations of Eu only the difference in intensity is occurred. With increasing Eu concentration TL intensity also get enhances and observed maximum for 1 mol%. Comparative TL glow curve show the concentration quenching shown in figure 9.Glow curve of CaSO4:Dy the phosphor which is widely used in TLD or personal dosimetry of ionizing radiations is also given for the comparison [5].

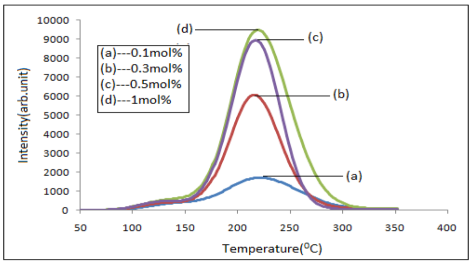

Fig.7- TL glow curves of $Na_2Sr_2Mg(BO_3)_2F_2:Eu^{3+}$ phosphor with different concentration of Eu^{3+} ions.

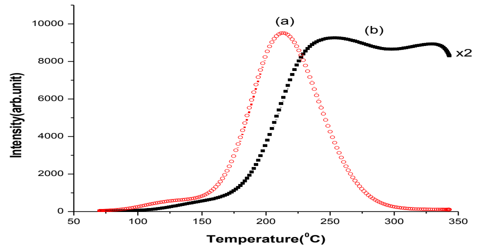

Fig. 8- Comparative TL glow curves of (a) $Na_2Sr_2Mg(BO_3)_2F_2:Eu^{3+}$ and (b) $CaSO_4$:Dy phosphors

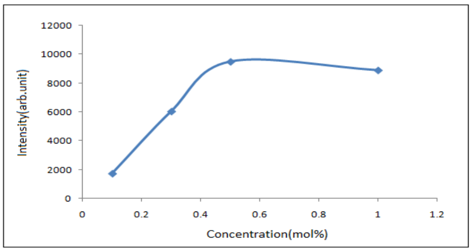

Fig. 9- Concentration of Eu^{3+} ions vs TL peak intensity graph of $Na_2Sr_2Mg(BO_3)_2F_2:Eu^{3+}$ phosphor

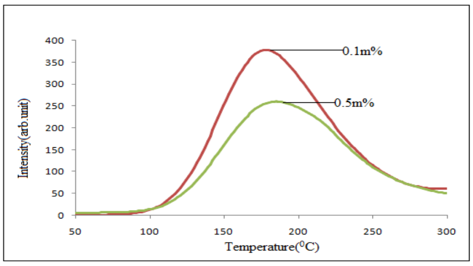

Fig. 10- TL glow curves of $Na_2Sr_2Mg(BO_3)_2F_2:Tb^{3+}$ phosphor with different concentration of Tb^{3+} ions

Fig 7 depict TL glow curve of Na2Sr2Mg(BO3)2F2:Tb3+ phosphor. Single glow curve with very less intensity is observed and promonent peak is seen at around 178ºC.

The intensity of the prominent glow peak.It is seen that TL intensity of $Na_2Sr_2Mg(BO_3)_2F_2$:Eu phosphor is 2 times less compared to standard $CaSO_4$:Dy phosphor.

Conclusions

PL characterization of $Na_2Sr_2Mg(BO_3)_2F_2$ phosphor doped with Tb shows a strong green emission located at 545 nm which is due to the transitions from the excited state $^5D_4$ to the ground.

States $^7F_5$ indicate that it could be a good green phosphor candidate for creating white light in phosphor converted white LEDs.

TL results of rare earth(RE=Tb,Eu) doped $Na_2Sr_2Mg(BO_3)_2F_2$ fluoroborate phosphor is estimated in this project report. From the results presented here, it is seen that powders of rare earth (RE=Tb,Eu) activated fluoroborates were prepared in one step using simple wet chemical route. In case of $Na_2Sr_2Mg(BO_3)_2F_2:Eu^{3+}$ phosphor TL intensity is 2 times less compared to standard $CaSO_4$:Dy TLD phosphor.The compound may prove to be good host for other activators as well for thermoluminescence dosimetry.

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