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Synthesis and luminescence properties of MSO_4:Eu (M=Ca, Ba) phosphors for radiation dosimetry

C. B. Palan[1*], A. O. Chauhan[2], N.S. Sawala[3], S. K. Omanwar[4]

[1]*Department of Physics, Bapumiya Sirajoddin Patel Arts , Commerce and Science College, Pimpalgaon Kale, Tq. Jalgaon (Jamod), Dist:-Buldhana, Maharashtra, India
[2]Department of Physics, VidyaBharati College, Amravati, Maharashtra, India
[3]Science and Humanities Department, Government Polytechnic, Arvi, India
[4]Department of Physics, Sant Gadge Baba Amravati University, Amravati, India
*(Corresponding author email: chetanpalan27@gmail.com)

Abstract

The goal of this work was to investigate the relevant dosimetric and luminescent properties of MSO_4:Eu (M=Ca, Ba) phosphor for radiation dosimentry. The structural property was studied through X-ray diffraction technique. Additionally, the photoluminescence (PL), thermoluminescence (TL) and optically stimulated luminescence (OSL) behaviors of MSO_4:Eu (M=Ca, Ba) phosphor was studied. The XRD pattern of MSO_4:Eu (M=Ca, Ba) phosphor fully matched with the International center for diffraction data (ICDD) file. The PL spectra of as-prepared MSO_4:Eu (M=Ca, Ba) phosphor showed characteristic emission in near-UV region, The TL glow curve of MSO_4:Eu (M=Ca, Ba) phosphors were consist overlapping peaks in temperature 50–300°C. Also MSO_4:Eu (M=Ca, Ba) phosphors show excellent CW-OSL response under β irradiation.

Keywords: Radiation dosimetry; Luminescence properties; X-ray diffraction; Thermoluminescence; Optically Stimulated Luminescence; MSO_4:Eu (M=Ca, Ba).

1. Introductions

Radiation dosimetric methods are used for the estimation of dose absorbed by radiation in a detector material. These methods are required for estimation of absorbed dose in various applications of radiation, such as personnel and environmental dosimetry, retrospective, accidental, dosimetry and medical applications of radiation. The use of Thermoluminescence (TL) as a method for radiation dosimetry of ionizing radiation has been established for many decades and has found many useful applications in various fields, such as personnel, environmental, medical, archaeological, geological dating and space dosimetry. Several high sensitive TL phosphor materials and thermoluminescent dosimeters (TLDs) are now commercially available in different physical forms. There are many commercial TLD systems which are being used for various dosimetric applications and even presently, TL is a popular technique in the field of radiation dosimetry, particularly in personnel monitoring [1-4].

Thermoluminescent materials based on rare earth (RE)-doped alkaline earth sulphates have been studied since 1970 [5]. Sulphates are known to be good Thermoluminescent materials.Sulphate based RE doped phosphors, particularly alkaline earth sulphates, because of its high sensitivity, ease of preparation and stability of response in adverse climates, have already been very popular for use in radiation dosimetry, using thermoluminescence [6]. The main characteristics of some Eu activated sulphate TL materials are tabulated in **Table 1.**

Table 1. The main characteristics of some Eu activated sulphate based TL material

S.N	Material	Z_{eff}	Glow peak Temp (°C)	Synthesis Method	Application	Ref.
1.	$BaSO_4$:Eu	30.3	190	Precipitation method	Radiation dosimetry using OSL as well as TL	[7]
2.	$BaSO_4$: Eu	30.3	-	Recrystallization method	Dosimetric applications .	[8]
3.	$SrSO_4$:Mn	46.5	346	Wet chemical precipitation method	High temperature radiation dosimetry applications	[9]
4.	$SrSO_4$:Eu	46.5	231	Acid evaporative method		[10]
5.	$SrSO_4$:Eu	46.5	312	Chemical precipitation method	Research in mixed fields detection dosimeters	[11]
6.	$SrSO_4$:DyTb	46.5	217	Co-precipitation method	Estimating high dose of gamma rays	[12]
7.	$CaSO_4$: Eu	15.3	180	-	TLD	[13]
8.	$CaSO_4$: Eu	15.3	173	Chemical co-precipitation method	High dose in space craft payload	[14]

In the last two decades an alternative technique namely OSL has been developed and widely used. The OSL technique is the best of all the known techniques for measurement of radiation exposuressince the out process does not involve problems of blackbody radiation and thermal quenching as in TL [15-18].

Many studies have searched for new OSL materials with suitable properties for dosimetry, including phosphate, borate, sulfate and halides [19-34]. Rare earth activated MSO_4 is an interesting material for TL/OSL, it has a low effective atomic number with hydrated Orthorhombic phases [35]. Rare earths (RE) activated sulfate based phosphors are widely used for radiation dosimetry.

The purpose of this work was to investigate the dosimetric and luminescence properties of a MSO_4:Eu (M=Ca, Ba) phosphor, this phosphor is developed via co-precipitation method for radiation dosimetry application. However, to the best of our knowledge, there are no reports on the Comparative studies of TL/OSL properties of MSO_4:Eu (M=Ca, Ba) phosphor.

2. Experimental details

MSO_4 (M=Ca, Ba) phosphors activated with Eu were prepared by co-precipitation method described in our earlier works [36]. The sitochometry of the reaction is maintained by formula $M_{1-x}SO_4:xEu^{2+}$(M=Ca, Ba). The nitrate precursor of Calcium/ Barium was dissolved in 100 ml of double-distilled water with addition of the stock solution prepared for Eu_2O_3.Solution in glass beaker under stirring to form a homogeneous aqueous solution and it was confirmed that precursor was dissolved in water. 10 ml H_2SO_4solution was added drop by drop into the mixed aqueous solution of $M_{1-x}(NO_3)_2$: xEu (M=Ca, Ba) under rigorous stirring at room temperature and white precipitation formed. After that, the MSO_4(M=Ca, Ba) precipitate was centrifuged and washed several times by water to remove the excess residual salts. The precipitate was dried at 60°C for 2 hr by optical heating. The dried sample was annealed at 900°C for 1 hr to get white crystalline powder of $MSO_4:Eu^{2+}$(M=Ca, Ba).The complete process involved in the reaction was represented as a flow chart in **Fig. 1.**

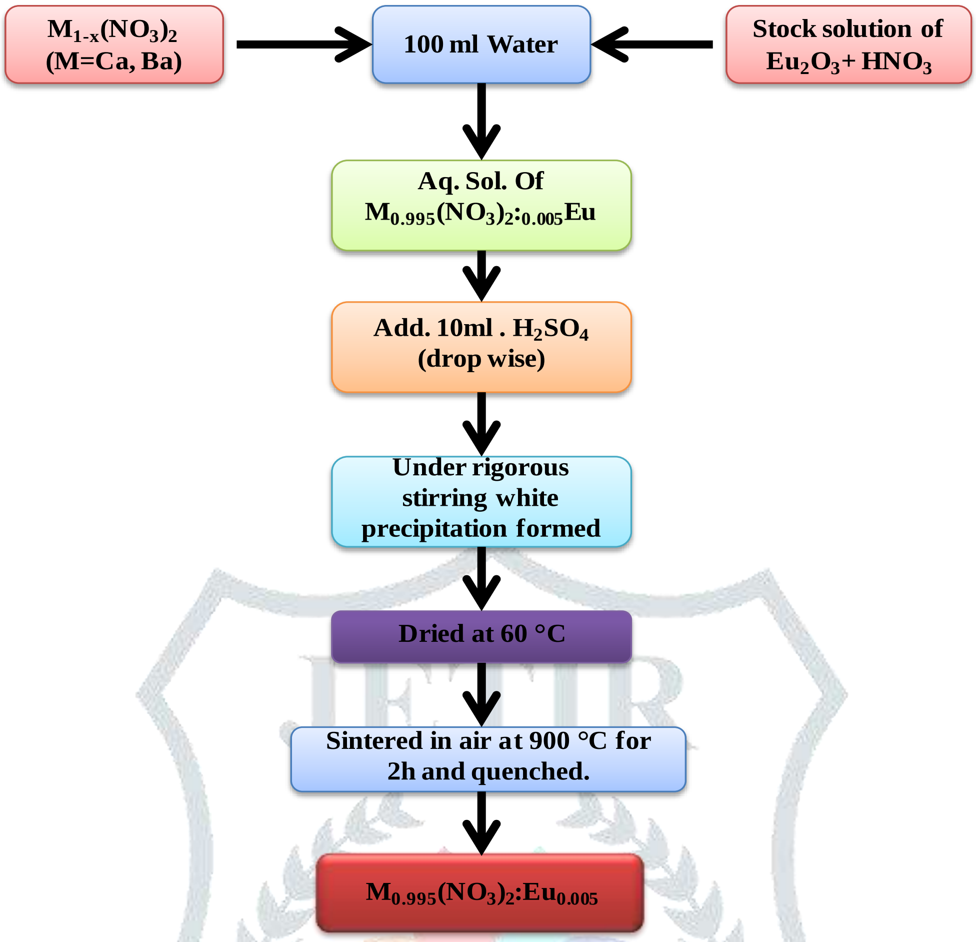

Fig. 1 Flow chart of $M_{(1-x)}$ SO_4:xEu^{2+}(M=Ca, Ba) synthesized by Co-Precipitation method.

3. Results and discussion

The structure of the as-prepared samples were analyzed by Rikagu Miniflex X-ray diffractometer, using monochromatic CuKα_1 (λ = 1.5405 Å) radiation in the 2θ range of 10-60°. Photoluminescence was studied on a Hitachi F-7000 fluorescence spectrophotometer. Emission and Excitation spectra were recorded using a spectral slit of 2.5 nm for each window. For studying the TL and OSL response, all the samples were irradiated using ^{90}Sr/^{90}Y beta source with the dose rate of 20mGy per minute. All OSL measurements were carried out using an automatic Risø TL/OSL-DA-15 reader system which can accommodate up to 48 discs at IGCAR . Blue LEDs emitting at 470 nm (FWHM = 20 nm) are arranged in four clusters each containing seven individual LEDs. The total power from 28 LEDs at sample position is w80 mW/ cm^2. A green long pass filter (GG-420) is incorporated in front of each blue LED cluster to minimize the amount of directly scattered blue light from reaching the detector system. The standard PMT used in the Risø TL/OSL luminescence reader is a bialkali EMI 9235QA, which has an extended UV response with maximum detection efficiency between 300 and 400 nm. To prevent scattered stimulation light from reaching the PMT, the Risø reader is equipped with a 7.5 mm Hoya U-340 detection filter, which has a peak transmission around 340 nm (FWHM ~80 nm)

3.1. *X-ray diffraction pattern*

The structure of MSO_4:Eu^{2+}(M=Ca, Ba) phosphor was orthorhombic.

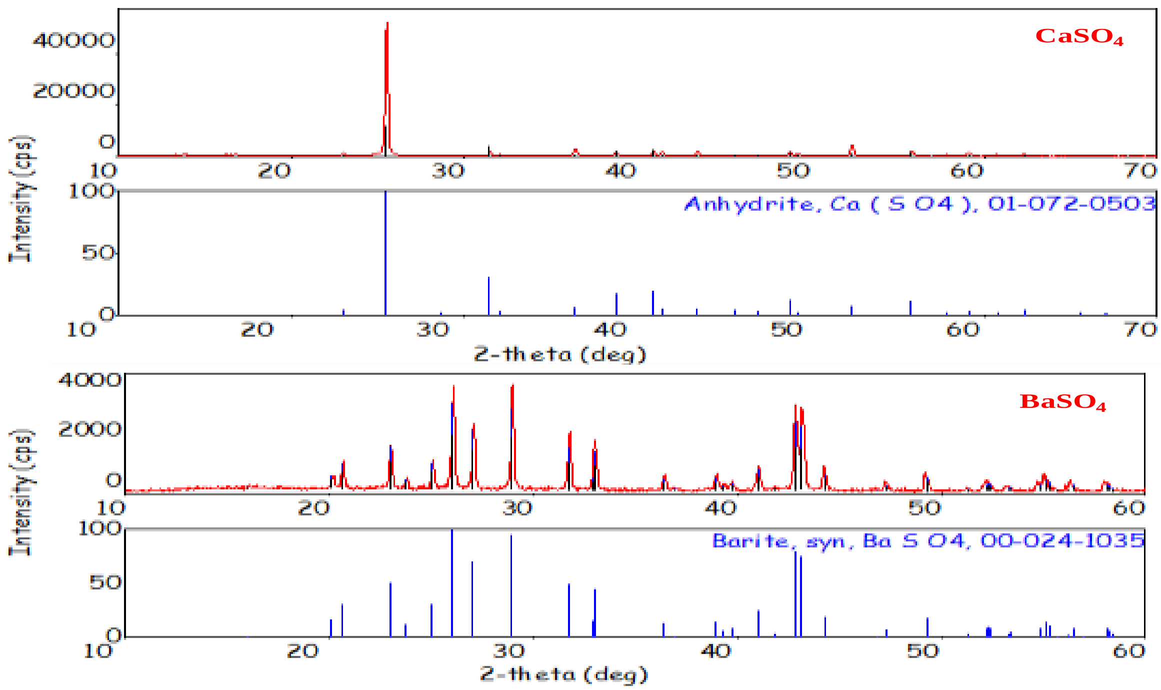

Fig. 2 X-Ray diffraction patterns of MSO_4:Eu(M=Ca, Ba)phosphors with ICDD patterns

In order to determine the phase purity, chemical nature of the phosphor, X-ray diffraction (XRD) analysis was carried out. **Fig. 2** show the XRD pattern of MSO_4:Eu^{2+}(M=Ca, Ba) phosphor along with the standard XRD pattern (ICDD Card No. 01-072-0503 and 00-024-1035). The XRD pattern shows the formation of pure $CaSO_4$ and $BaSO_4$ phase. The addition of the dopant (Eu) does not seem to have effect on the XRD pattern which suggests that the dopant was incorporated in the lattice.

3.2 Photoluminescence Properties (PL)

The combine excitation and emission spectra of MSO_4: Eu (M=Ca, Ba) phosphors were give in **Fig. 3.** The excitation and emission spectra of $BaSO_4$:Eu phosphor was observed at 377 nm and 326 nm respectively. The excitation and emission spectra of $CaSO_4$:Eu phosphor was observed at 384 nm and 320 nm respectively.

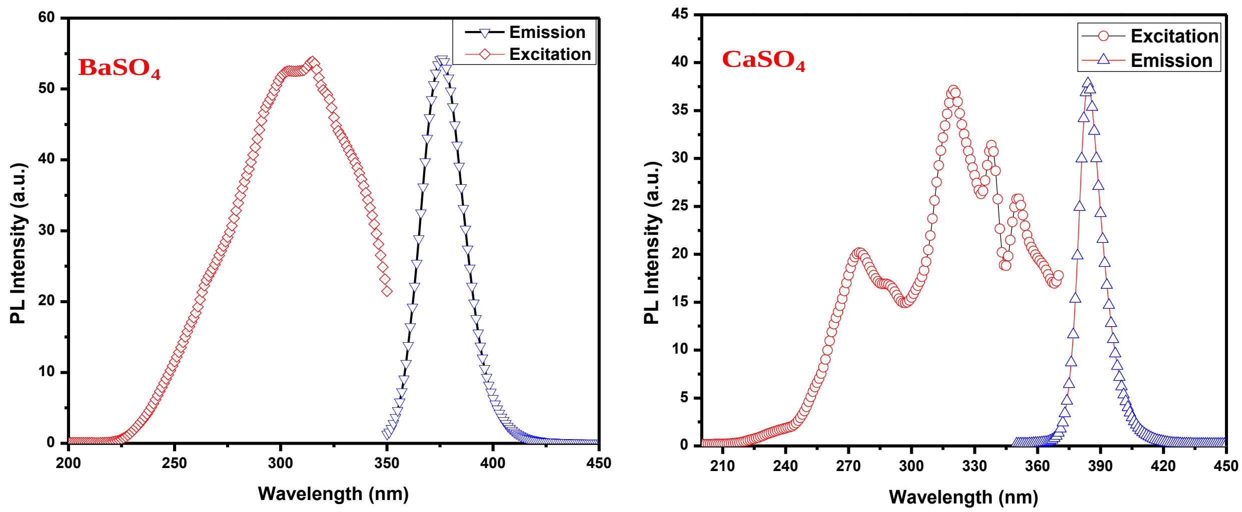

Fig. 3 Excitation and emission spectra of MSO_4: Eu (M=Ca,Ba) phosphors

3.3 Thermoluminescence (TL)

Thermoluminescence glow curve of MSO_4: Eu (M=Ca, Ba) phosphors under β irradiation as shown in Fig. 4. The TL glow curve of the MSO_4: Eu (M=Ca, Ba) phosphor consists of overlapping peaks in temperature rang 50-300°C. The TL sensitivity of $BaSO_4$:Eu phosphor was 5 time sensitive than TL sensitivity of $CaSO_4$:Eu phosphor.

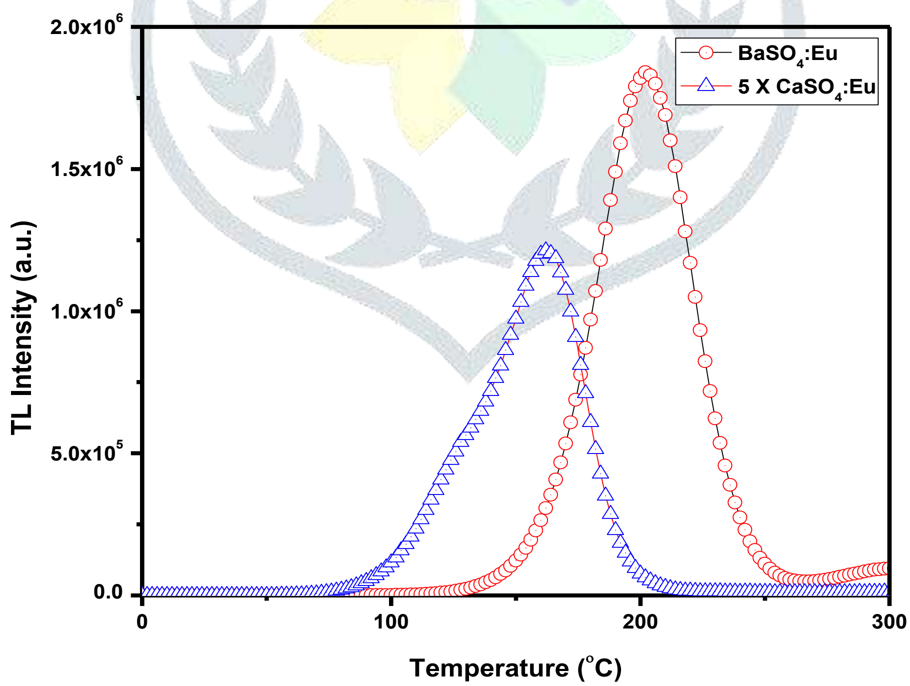

Fig. 4 Comparison of TL responses MSO_4: Eu (M=Ca,Ba) phosphors under beta irradiation

3.4 Optically stimulated luminescence

The CW-OSL response of prepared MSO_4: Eu (M=Ca, Ba) phosphor under 100mGy of β irradiation as shown in **Fig. 4.** The OSL sensitivity of compared with two different [37]. In the first method, The OSL sensitivity of $BaSO_4$:Eu phosphor was found to be about 40 times to that of $CaSO_4$:Eu phosphor. OSL sensitivity of $CaSO_4$:Eu phosphor was more than $BaSO_4$:Eu phosphor by using second method. The difference in the sensitivities in the two methods is because of the fact that the OSL decay in the $BaSO_4$:Eu based phosphors is faster as compared to $CaSO_4$:Eu phosphor.

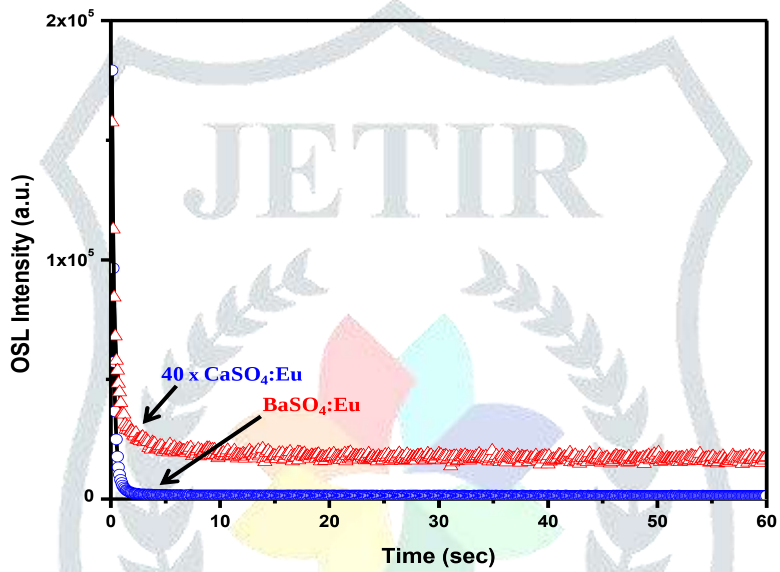

Fig. 4 Comparison of OSL responses MSO_4: Eu (M=Ca,Ba) phosphors under beta irradiation

4. Conclusions

In this reports MSO_4: Eu (M=Ca,Ba) phosphor was synthesized via Coprecipitaion method. The XRD pattern of prepared MSO_4: Eu (M=Ca,Ba) phosphor was perfectly matched with ICDD file. The TL glow curve consist overlapping peaks in temperature 50-300°C range. The emission spectra of prepared phosphor was obsreved in 350 – 450 nm range. The CW-OSL decay pattern of prepared MSO_4: Eu (M=Ca,Ba) phosphor was same than CW-OSL decay pattern of commercial available α-Al_2O_3:C phosphor. The OSL sensitivity of $BaSO_4$:Eu phosphor was found to be about 40 times to that of $CaSO_4$:Eu phosphor. The OSL decay in the $BaSO_4$:Eu based phosphors is faster as compared to $CaSO_4$:Eu phosphor. The effective atomic number of prepared MSO_4: Eu (M=Ca,Ba) phosphor is nearly s 15. Although MSO_4: Eu (M=Ca,Ba) is not a material equivalent to tissue but, due to its high TL and OSL sensitivities, this phosphor can be proposed as a suitable candidate for radiation dosimetry, of course, after further progress in the studies.

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