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Effect of gamma irradiation on thermoluminescence and fracto-mechanoluminescence properties of $SrMgAl_{10}O_{17}:Eu^{2+}$ phosphor

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$SrMgAl_{10}O_{17}:Eu^{2+}$ phosphor has been synthesized by combustion method using urea as a fuel. Thermoluminescence (TL) and mechanoluminescence (ML) properties of synthesized phosphors under gamma irradiation were reported and discussed in this paper. The structural and morphological studies were done using X-ray diffraction (XRD) and scanning electron microscopy (SEM). Photoluminescence emission spectrum is obtained at 460 nm. Thermoluminescence glow curves of synthesized phosphor show a broad peak, which has been deconvoluted into three peaks and activation energies were calculated using peak shape method. Total mechanoluminescence (ML) intensity increases linearly with gamma doses.

1. Introduction

Luminescence materials have attracted significant attention to meet the increasing requirement in lighting and display fields. Various aluminates are used as host for doping rare earth ions in luminescent applications. The Eu^{2+} doped barium and strontium magnesium aluminates, $MMgAl_{10}O_{17}:Eu^{2+}$ (M = Ba, Sr), are among the recent commercially exploited luminescent materials [1–7]. Most recently, Xing et al. [8] have proposed that $SrMgAl_{10}O_{17}$:Eu^{2+} prepared by high temperature solid state reaction would be a promising blue phosphor material for PDP application.

Rare-earth doped luminescent materials play a significant role as radiation detectors in many fields, for example nuclear power plants, radiotherapy, personal and environmental monitoring of ionizing radiation and in geological dating. Thermoluminescence (TL) is the emission of light beyond thermal equilibrium, subsequently occurring after the absorption of energy from an external source. In recent years thermoluminescence properties of rare earth doped aluminate based phosphors have been reported by several workers [9,1,10–13]. There have been no previous published reports on the thermoluminescence of gamma irradiated $SrMgAl_{10}O_{17}:Eu^{2+}$ phosphor so we have attempted to study the thermoluminescence property in order to find its suitability for dosimetric applications.

The cold light emission due to fracture of solids is called fracto-mechanoluminescence (FML). In the recent past, the FML phenomenon has attracted the attention of a large number of researchers all over the world because of its potential application to damage sensor of structures. FML has a great deal of potential to understand the following facts and devices: (i) earthquake and mine failure [14], (ii) earthquake lights [15,16], (iii) design of damage sensor [17,18], (iv) design of fracture sensor [19–21] etc. Mechanoluminescence of rare earth doped aluminates have been studied extensively [22–25]. Morito Akiyama et al. [26] investigated stress induced mechanoluminescence (ML) from $SrMgAl_6O_{11}$:Eu and reported that $SrMgAl_6O_{11}$: Eu phosphor emits the highest mechanoluminescence intensity among $Sr_3Al_2O_6$:Eu, Dy, $SrAl_2O_4$:Eu, $Ca_2Al_2SiO_7$:Ce and ZrO_2:Ti phosphors. We have investigated the ML properties of $SrMgAl_{10}O_{17}:Eu^{2+}$ phosphor and found that this phosphor exhibits strong ML. To the best of our knowledge FML properties in $SrMgAl_{10}O_{17}:Eu^{2+}$ phosphor has not been investigated earlier. So it is planned to study the ML properties in gamma irradiated $SrMgAl_{10}O_{17}:Eu^{2+}$ phosphor. The dependence of ML intensity on different gamma doses and on different impact velocities were investigated to find its possible application in ML dosimetry.

2. Experimental details

Analytical grade strontium nitrate Sr $(NO_3)_2$, aluminium nitrate Al $(NO_3)_3$, magnesium nitrate Mg $(NO_3)_2$, europium oxide Eu_2O_3 and urea CO $(NH_2)_2$ were used as the starting raw materials. These powders were weighted according to their stoichiometry in the

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normal composition of $SrMgAl_{10}O_{17}:Eu^{2+}$. In order to form Eu $(NO_3)_3$, Eu_2O_3 was dissolved in 2 ml dilute HNO_3 solution. Then weighed quantities of each nitrate and urea were mixed together and crushed into mortar for 1 h. The resulting paste was transferred to crucible and kept in a muffle furnace maintained at 600 °C initiating temperature. Initially the mixture boils and undergoes dehydration followed by decomposition with the evolution of the large amount of gases (oxides of carbon, nitrogen and ammonia). The process is highly exothermic continues and the spontaneous ignition occurs. The solution underwent smoldering combustion with enormous swelling, producing white foamy and voluminous ash. The foamy product can easily be milled to obtain the precursor powder. The phase analysis of synthesized sample was carried out by PANalytical 3 Kw X'pert Powder-Multifunctional XRD instrument. The surface morphology of prepared phosphor and Energy Dispersive X-ray (EDX) spectra was analyzed by ZEISS EVO Series Scanning Electron Microscope Model EVO 18. Photoluminescence (PL) emission and excitation spectra were measured using Shimadzu RF5301PC spectroflurophotometer. The TL glow curves were recorded by Nucleonix TL 1009I. Mechanoluminescence studies are done using the homemade indigenous setup where RCA-931 PMT kept below the lucite plate and interfaced to a storage oscilloscope (Scientific 300 MHz, SM340) [27]. ML and TL spectra were recorded using interference filters of different wavelengths.

3. Results and discussion

3.1. X-ray diffraction analysis

The XRD pattern of synthesized sample is shown in Fig. 1. The diffraction peaks of $SrMgAl_{10}O_{17}$: Eu (SAM: Eu) phosphor matched well with JCPDS Card No: 26-0879. The crystal structure of SAM is hexagonal and belongs to the space group $P6_3/mmc$.

3.2. Morphology analysis

The SEM images of $SrMgAl_{10}O_{17}:Eu^{2+}$ (1 mol%) are shown in Fig. 2. It is seen that in SEM images the surface of prepared phosphor has pores. These pores are formed when gas escapes under high pressure in the combustion process. The non-uniform and irregular shapes of the particles as shown in Fig. 2 may be attributed to the non-uniform distribution of temperature and mass flow in the combustion flame [28,29].

Fig. 3(a) shows the Energy Dispersive X-ray (EDX) spectra of $SrMgAl_{10}O_{17}:Eu^{2+}$ (1 mol%) phosphor. The EDX spectra confirm the elemental composition of synthesized phosphor. Table 1 shows

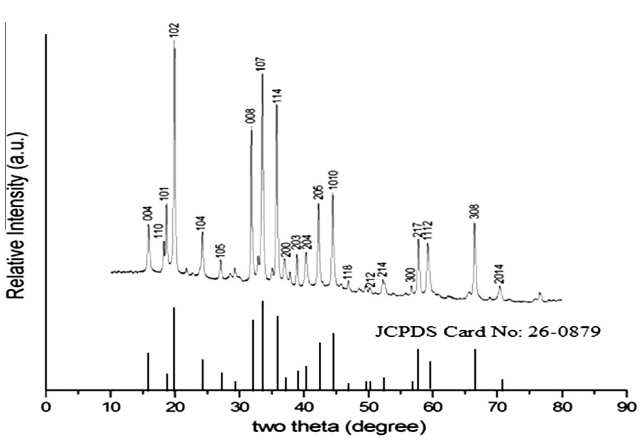

Fig. 1. XRD pattern of $SrMgAl_{10}O_{17}:Eu^{2+}$ (1 mol%) phosphor.

the quantitative amount of the elements present in the synthesized phosphor in a specific area indicated in the SEM image of phosphor shown in Fig 3(b).

3.3. Photoluminescence study

Fig. 4 shows the excitation and emission spectra of $SrMgAl_{10}O_{17}:Eu^{2+}$ (1 mol%) phosphor. The excitation spectra exhibit a peak centered at 340 nm. The PL emission spectra of $SrMgAl_{10}O_{17}:Eu^{2+}$ (1 mol%) phosphor has one broad peak around 460 nm due to the transition $4f^65d^1 \rightarrow 4f^7$ of Eu^{2+} ions as shown in Fig 4. Fig. 5 shows the CIE diagram of $SrMgAl_{10}O_{17}:Eu^{2+}$ (1 mol %) phosphor. The chromaticity coordinates of $SrMgAl_{10}O_{17}:Eu^{2+}$ (1 mol%) phosphor falls in the blue region.

3.4. Thermoluminescence studies

Thermoluminescence (TL) is used for characterization of traps and recombination centers in insulating or semiconductor materials. Commercial applications of thermoluminescence include dosimetry of ionizing radiation and dating of archeological and geological objects. For the measurement of thermoluminescence (TL) a sample is pre excited by UV, gamma, X-ray etc. in order to fill traps and recombination centers with charge carriers. Then, the sample is heated, usually with a constant rate (in TL). The luminescence is related to radiative recombination of charge carriers, which were thermally or optically released from traps and then moved to recombination centers. Thermoluminescence (TL) glow curves of $SrMgAl_{10}O_{17}:Eu^{2+}$ phosphors for different concentrations of Eu were recorded by thermoluminescence reader TL 1009I, with a heating rate of 5 °C/s. For the Thermoluminescence measurement, samples were irradiated with γ-rays using a ^{60}Co source having an exposure rate of 590 Gy/hr and the TL response was observed for different gamma doses.

Fig. 6 shows the dependence of total TL intensity on the doping concentration of Eu in $SrMgAl_{10}O_{17}$ host. Total TL intensity found maximum for 1 mol% concentration of Eu. Further increase in Eu concentration reduces the total TL intensity due to concentration quenching. Fig. 7 represents the TL glow curves of $SrMgAl_{10}O_{17}$: Eu^{2+} (1 mol%) phosphor for different gamma irradiation doses. It is observed that TL intensity increases with gamma doses. The increase in TL intensity with increasing gamma dose is due to increase in density of trapped carriers. Fig. 8 shows the total TL response of synthesized phosphor as a function of gamma dose. A linear relationship between gamma dose and total TL intensity is observed as shown in Fig. 8. This linearity property is one of the essential properties of a phosphor to be used as TL dosimeter for ionizing radiation. For dosimetric purpose the TL response of phosphor should stable and the fading must be a minimum upon storage after being exposed to ionizing radiation. In order to check the suitability of synthesized phosphor as dosimeter the fading analysis is done. The TL fading of $SrMgAl_{10}O_{17}:Eu^{2+}$ (1 mol%) phosphor was recorded over 90 days which is shown in Fig. 9. We found that the fading is less. The thermoluminescence spectra of $SrMgAl_{10}O_{17}:Eu^{2+}$ (1 mol%) phosphor found at 460 nm as shown in Fig. 10.

3.5. Analysis of the TL glow curves

Computerized glow curve deconvolution (CGCD) analysis has been applied to resolve a complex thermoluminescence glow curve of $SrMgAl_{10}O_{17}:Eu^{2+}$ phosphor into individual peak components and the trapping parameters such as activation energy and frequency factor are evaluated.

The method used for the calculation of activation energy