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Synthesis and thermoluminescence properties of $SrAl_2O_4$ (EU) phosphor irradiated with cobalt-60, 6 MV and 16 MV photon beams

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HIGHLIGHTS

- $SrAl_2O_4$(Eu) phosphor was synthesized by low temperature combustion technique.
- Average crystalline size was found to be ~33.04 nm.
- Thermoluminescence was studied after irradiation with Co-60, 6 and 16 MV photons.
- Glow peaks obey second order kinetics and have 0.784 eV of activation energy.
- Phosphor can be utilized for dating, temperature sensor and radiation dosimetry.

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ABSTRACT

Powder samples of $SrAl_2O_4$ (Eu) were synthesized by the combustion method using urea as a fuel. The combustion products were calcined at 700 °C for 1 h. X-ray diffraction (XRD) patterns of the prepared sample exhibit sharp diffraction peaks and absence of any amorphous phase. The average crystalline size was found to be ~33.04 nm, calculated by using Debye Scherer's formula. The scanning electron microscope (SEM) images reveal that the crystallites have no uniform shape and the presence of several micro- and nano-particles within the grain. This may be due to the non-uniform distribution of temperature and mass flow in the combustion flame which results in the non-uniform shape of crystallites. The thermogravimetric analysis (TGA) indicates that the prepared sample is thermally stable up to 900 °C. Thermoluminescence (TL) behavior of prepared samples was studied after irradiation with Co-60gamma rays, 6 mega voltage (MV) and 16 MV photon beams at various doses. Glow curve of the prepared $SrAl_2O_4$ (Eu:1%) sample was similar in shape irrespective of incident energy and radiation type. The dominant peak in each glow curve appeared around at 312 °C. No shifts in peak positions have been observed. All the glow curves of sample doped with Eu(3%) have relatively higher intensity as compared to the sample doped with Eu(1%). Energy dependence has been observed in the present phosphor. This could be because of increase in the probability of Compton's interaction at this energy range due to transmission of primary as well as scattered radiation and decrease in mass attenuation coefficient with the increase in energy. The trapping parameters namely activation energy (E), order of kinetics (b) and frequency factor (s) have been determined using the glow curve shape (Chen's) method. These phosphors could be utilized for display applications, dating, temperature sensor, low as well as high energy radiation detection and dosimetry especially where tissue equivalency is not much desirable like gamma irradiator, environmental and retrospective dose assessment.

1. Introduction

Luminescence is a common phenomenon among inorganic and organic insulators as well as in semi-conductors. The long lasting luminescence, also called phosphorescence, is a potential candidate for the various applications such as in display technology, warning signal lights, luminous paints for highways, detection of particulate and non-particulate radiations, archeological dating, temperature sensor, determination of radiation absorbed dose, exposures, etc. (McKeever, 1985; Daniels et al., 1953; Furetta, 2003). Strontium aluminates based phosphors have attracted much attention for their high quantum efficiency, long lived afterglow, good chemical stability and other luminescence related

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features (Matsuzawa et al., 1996; Ayvacikli et al., 2011; Xiao et al., 2010; Arellano-Tanori et al., 2008; Sharma et al., 2009). There are a number of synthesis techniques to prepare phosphor for the above said applications. Few among these are solid state reaction method, sol–gel method, precipitation method (Xiao et al., 2010) and combustion method (Patil, 1993). Solid state reaction method requires high temperature for long duration, sol–gel and precipitation methods require comparatively low temperature but more chemistry involved. The combustion technique needs low temperature as well as fewer hours to accomplish.

Phosphors produced with solid state reaction method generally have larger particle size as compared to combustion produced, as a result decrease in luminescence intensity has been reported (Xiao et al., 2010). The nanophosphors have high surface to volume ratio and show the changes in surface electronic states (Premkumar et al., 2013). A high concentration of surface atoms and defects at multiple nanograin boundaries may be regarded as one of the fundamental properties of nanophosphors (Kortov, 2010). The nanophosphors have high radiation resistance as compared to microphosphors and their bulk analog. This raises the upper limit of dose measurement. This also enables the nanophosphors to use for measurement of intensive flow of high energy ions, high power X and gamma radiation fields, skin dose etc. In nanophosphors the dosimetric peak is shifted towards high temperature side, hence fading can be minimized (Kortov, 2010, 2007). To synthesize the advanced nanophosphors low temp combustion route has been used widely. In combustion method exothermic chemical reaction takes place for a few seconds and the whole process is completed within 5 min. It is a highly time-saving approach to prepare the powder phosphors with desired phase and purity. The thermoluminescence (TL) or thermally stimulated luminescence (TSL) is the release of trapped energy in the form of ultra-violet (UV) or visible light. In this process heat is the stimulating agent. The release of trapped energy is proportional to the absorbed dose in the phosphor materials (McKeever, 1985; Kortov, 2007; Bos, 2007; Pawel, 2010).

When a solid is irradiated with ionizing radiations, a partial or full, energy of the incident beams is absorbed in the material. Electrons and holes are created and subsequently they migrate and are trapped at the defect sites available in the phosphor material. When the irradiated phosphor is heated the electrons and holes release from their respective trapped sites and re-combine with their counter parts; as a result annihilation takes place in the form of phosphorescence. Polycrystalline phosphors exhibit a glow curve, which is thermoluminescence intensity versus temperature plot, with single or multiple peaks (Berezovskaya et al., 2007; Manam and Das, 2010; Sharma et al., 2010; Ranogajee-Komor and Osvay, 1986; Murthy et al., 2006; Hunter et al., 2012). The glow curve provides the TL parameters as activation energy (E) frequency factor (s) and order of kinetics (b) (Chen, 1969; Li et al., 2005; Chandrasekhar et al., 2012; Sangeeta, 2004). Because the TL intensity is related to the absorbed radiation dose in the material, the thermoluminescence is widely used in radiation dosimetry and geological dating (McKeever, 1985; Furetta, 2003). Application of thermoluminescence phenomenon for dosimetric purposes has been started since 1953 (Daniels et al., 1953; Manam and Das, 2010). There has been a constant need for high sensitive TL phosphors that can be used for vast applications. The TL method of radiation monitoring is one of the most economical, easy to achieve and readily adaptable radiation measuring technique for any laboratory. In the present work we have investigated the TL properties of combustion synthesized nanophosphor after irradiation with gamma (Co-60) and mega-voltage (6 and 16 MV) photon beams.

2. Materials and methods

2.1. Synthesis

Combustion synthesis was used for the preparation of strontium aluminate ($SrAl_2O_4$) phosphor doped with europium (Eu). The stoichiometric amounts of ultra pure and analytical (AR) grade $Sr(NO_3)_2$, $Al(NO_3)_3 \cdot 9H_2O$, $Eu(NO_3)_3 \cdot 6H_2O$ (Biochemika, Otto) and urea (NH_2CONH_2) from s.d.fiNE-Chem Ltd. were calculated according to the total oxidizing and reducing valence of the components such that the equivalence ratio (O/F) should be unity and the energy release is maximum (Patil, 1993). Here the metal nitrates act as oxidizer (O) and urea as a fuel (F). All the starting materials were thoroughly mixed in an agate mortar using minimum amount of de-ionized water. The resulting homogeneous thick paste was transferred in a borosilicate beaker of 500 ml and placed in the muffle furnace which was pre-heated at around 650 °C. Initially the paste boils and undergoes dehydration, followed by decomposition and evolution of gases. The mixture then froths and swells forming foam, which ruptures with a flame and glows to incandescence. During incandescence the foam further swells to the capacity of container. The entire combustion process was over within 5 min. The pre-calcined product was obtained after crushing and thoroughly mixing the combustion product in an agate mortar for 10 min. Prepared powder was then transferred into a silica crucible and inserted in the furnace for calcination at 700 °C for 1 h. After calcinations the powder was again thoroughly mixed in an agate mortar to homogenize the final product. The obtained final powder sample was then stored in dark for further studies.

2.2. Characterization

To confirm the phase formation and evaluation of crystallites of the prepared sample, X-ray diffraction (XRD) study was performed using PANalytycal Empyrean Diffractometer with Cu-Kα ($\lambda = 1.541$ Å) radiation with a nickel filter. The surface morphology of the sample was examined by using JEOL/EO JSM-6390 scanning electron microscope (SEM). Thermogravimetric analysis (TGA) was carried out to check the thermal stability of prepared powder sample using Mettler Toledo (TGA/DSC1) instrument. TGA analysis of the sample was carried out in air atmosphere at the heating rate of 10 °C/min and taking $\approx$ 11 mg sample weight before calcinations. Samples were irradiated with 50, 100 and 200 Gy by Co-60 source (gamma rays of 1.25 MeV (average) energy) installed in teletherapy machine (Theratronics 780C, Canada) with the dose rate of 1.8 Gy/min for the standard (10 cm × 10 cm) field size and depth. Samples were also irradiated for 100 Gy with high energy X-ray photons (6 MV and 16 MV) of linear accelerator (2300C/D, Varian, Palo Alto, USA) with the dose rate of 300 MU/min. The average energy of the 6 MV and 16 MV X-ray photons is approximately 1.5 MeV and 5 MeV respectively. Build-up material with appropriate thickness was applied over the phosphor sample at the time of irradiation to deliver the desired dose in the sample. The doses were verified by using the reference class 0.6 cm^3 ion chamber (FC-65G, IBA Dosimetry Germany) calibrated against the primary standard at national calibration laboratory (BARC, India). The dose measurement was done according to International Atomic Energy Agency (IAEA) TRS-398 protocol (TRS-398, 2000).

Approximately 3 mg $\pm$ 5% of powder sample was taken for TL measurement in each case. TSL measurements were performed using a thermoluminescence (TLD) reader (Harshaw 3500) in the temperature range between 50 and 400 °C with the linear heating rate of 2 °C/s. It is a planchet based (Ohmic) heating reader. It has heating temperature capability up to 400 °C. The thermoelectric PMT cooler has been used for maximum gain stability. The PMT