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Combustion synthesis of rare earth activated and co-activated $SrAl_4O_7$ green long lasting phosphors

A.N. Yerpude [a,*], S.J. Dhoble [b]

[a] Department of Physics, N.H. College, Bramhapuri, Chandrapur 441206, India
[b] Department of Physics, RTM Nagpur University, Nagpur 440033, India

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ABSTRACT

Green emitting $SrAl_4O_7:Eu^{2+}$, $SrAl_4O_7:Eu^{2+}$, Dy^{3+}, $SrAl_4O_7:Eu^{2+}$, Dy^{3+}, Ce^{3+} phosphors were synthesize by combustion method. The phosphor samples were well characterized by X-ray diffraction (XRD), Scanning Electron Microscopy (SEM), Photoluminescence(PL) emission and excitation spectra, Fourier transform infrared (FTIR) study, luminescence decay and afterglow decay. The excitation and emission spectra indicates that the phosphor could be well excited by UV light and give broad band emission due to the $4f^65d \rightarrow 4f^7$ transition of Eu^{2+} ions. All the phosphors show green long lasting phosphorescence phenomena after the excitation source was removed. The $SrAl_4O_7:Eu^{2+}$, Dy^{3+}, Ce^{3+} phosphor has longest afterglow comparably than $SrAl_4O_7:Eu^{2+}$ and $SrAl_4O_7:Eu^{2+}$, Dy^{3+} phosphors. The $SrAl_4O_7:Eu^{2+}$, Dy^{3+} phosphor exhibits one thermally stimulated luminescence (TSL) peak at 90 °C and $SrAl_4O_7:Eu^{2+}$, Dy^{3+}, Ce^{3+} phosphor exhibits peak at 99 °C. The luminescence decay curves are well fitted with double exponential equation. Scanning electron microscopy (SEM) study was carried out to investigate the surface morphology and the crystallite sizes.

1. Introduction

Long lasting phosphors are kind of energy-storing material which can absorb energy and then gradually emits visible light for a decay time extending to seconds, minutes or even hours at room temperature after removal of the excitation [1–4]. The persistent luminescence materials have attracted a lot of attention in recent years owing to their potential practical applications such as emergence lighting displays, detection of high energy rays, multidimensional optical memory, luminous paints, safe traffic, wall painting, safety indication, fingermark detection and emergency lighting and imaging storage [2,5,6]. The photoluminescence of Eu^{2+} and Dy^{3+} co-doped alkaline earth aluminates phosphor has been widely studied because these phosphors have satisfactory luminescent properties [7–11]. A particular feature of these compounds is that when another trivalent rare-earth ions such has (e.g. Nd^{3+}, Dy^{3+}, Ce^{3+}) is incorporated into the system, decay profile of the Eu^{2+} luminescence is altered. In europium doped phosphors the phosphorescence is observed which is ascribed to the allowed electronic transition of $4f^65d^1 \rightarrow {}^8S_{7/2}$ of the Eu^{2+} ion, which is strongly influenced by the host lattice. The emission of Eu^{2+} ions in various hosts can be varied from blue to red depending on the host lattice and synthesis methods [12,13]. Although many papers on various kinds of LLP phosphors have been published, the progress in developing commercial LLP phosphors is still rather slow. In this paper we report the combustion synthesis of $SrAl_4O_7:Eu^{2+}$, $SrAl_4O_7:Eu^{2+}$, Dy^{3+}, $SrAl_4O_7:Eu^{2+}$, Dy^{3+}, Ce^{3+}. These phosphors were well characterized by X-ray powder diffraction (XRD), scanning electron microscopy (SEM), Fourier transform infrared (FTIR), Photoluminescence(PL) measurement, Thermoluminescence(TL), Phosphorescence lifetime and afterglow decay.

2. Experimental procedure

2.1. Synthesis of phosphor

The $SrAl_4O_7:Eu^{2+}$(After we denote SAE), $SrAl_4O_7:Eu^{2+}$, Dy^{3+},(After we denote SAED), $SrAl_4O_7:Eu^{2+}$, Dy^{3+}, Ce^{3+}(After we denote SAEDC), phosphors were synthesized by a combustion method. Small quantities of B_2O_3 were used as a flux, amounts of urea were added as reducer and fuel, and metal nitrates were used as oxidizer. Metal nitrates were weighed stoichiometrically, dissolved in distilled water in a beaker and stirred in order to obtain a clear solution. The procedure used to prepare $SrAl_4O_7:Eu^{2+}$ had the following stages. First, $Al(NO_3)_3{\cdot}9H_2O$, $Sr(NO_3)_2$, B_2O_3

* Corresponding author. Tel.: +91 9637782611.
E-mail address: atulyerpude@gmail.com (A.N. Yerpude).

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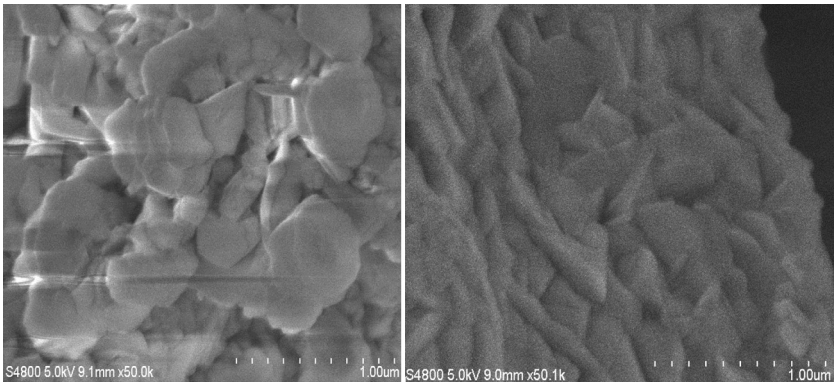

Fig. 1. Morphology of the $SrAl_4O_7$ phosphor.

and $CO(NH_2)_2$ were dissolved enough into distilled water to obtain transparent solution. Eu_2O_3 were dissolved into concentrated nitric acid to form nitrate solution, then mixed the two solutions together. The resulting colorless solution was further heated with stirring at 85–90 °C until a highly viscous wet gel was formed. The resulting solution was transferred into a china crucible, which was then introduced into a muffle furnace maintained at 550 °C. Initially, the wet gel boiled, followed by decomposition with the evolution of large amounts of gases (oxides of carbon and nitrogen). Then, spontaneous ignition occurred and underwent combustion with enormous swellings occurring. The whole process spent a few minutes and resulted into a foamy white powder. The entire combustion process being highly exothermic continues and the liberated gases swell the mixture into large volume. Large exothermicity results into a flame changing the mixture into gaseous phase. Flame temperature as high at 1600 °C converts the vapor phase into mixed aluminates. The flame persists for about 30 s. The voluminous and foamy combustion ash was milled to obtain the final $SrAl_4O_7$: Eu^{2+} phosphor. The final product obtained is in the fluffy form, which is used for the further investigations. Some corresponding methods were used to characterize the prepared phosphor.

2.2. Characterization of phosphor

The prepared host lattice was characterized for their phase purity and crystallinity by X-ray powder diffraction (XRD) using PAN-analytical diffractometer (Cu-*K*α radiation) at a scanning step of 0.01°, continue time 20 s, in the 2θ range from 10° to 80°. The morphologies of the products were examined by scanning electron microscopy (SEM, JEOL 6380A). The photoluminescence measurement of excitation and emission were recorded on the Shimadzu RF5301PC Spectrofluorophotometer. The same amount of sample 2 g was used for each measurement. Emission and excitation spectra were recorded using a spectral slit width of 1.5 nm. Phosphorescence lifetime spectrometer (Edinburgh Instruments, UK) using a μs Xe flash lamp. The LLP decay was measured by using a spectrophotometer (Perkin-Elmer LS 55). The thermoluminescence was recorded with the help of TLD reader (Nuclonix) in which the heating rate was 2 °C/min.

3. Results and discussion

3.1. XRD phase analysis

The formation of crystalline phases in the samples prepared by the combustion method was confirmed by powder XRD measurements. The XRD pattern of the prepared $SrAl_4O_7$ phosphor is well matched with standard JCPDs (Joint Committee on Powder Diffraction Standards) file no 00-025-1208, recently we have reported by the same method [14].

3.2. Morphology of the $SrAl_4O_7$ phosphor

Scanning electron microscopy (SEM) study was carried out to investigate the surface morphology of prepared phosphor as shown in Fig. 1. The micrographs show that the crystallite sizes are varying from few microns to several tens of microns. The particles possess foamy like morphology formed from highly agglomerated crystallites.

3.3. Fourier transform infrared (FTIR) study

Fig. 2 shows FTIR spectrum of prepared $SrAl_4O_7$ phosphor. FTIR is a useful technique for studying the molecular environment and nature of bonding of inorganic compounds. The band between 1500 cm^{-1} and 1700 cm^{-1} are assigned to the asymmetric vibration of —CH_2 group [15]. The bands that appear in the range 830–500 cm^{-1} are assigned to metal–oxygen stretching vibrations [16]. While the band appears at 1453 cm^{-1} is mainly due to Al—O vibrations. Sharp peak is observed at 1024 cm^{-1}, which is due to the Al—O vibrations in the spinel block of b-alumina structure having symmetry A_{2u} [17].

3.4. Photoluminescent properties of the SAE, SAED, SAEDC phosphor

The excitation and emission spectra of SAE, SAED and SAEDC are shown in Figs. 3 and 4. Emission spectra of SAE, SAED and SAEDC phosphors show broad band emission, which is the transition of Eu^{2+} that originates from $4f^6 5d$ excitation state to $4f^7$ ground state. The SAE phosphor show blue-green emission band centered at 491 nm, which is attributed to the typical $4f^6 5d^1 \rightarrow 4f^7$ transition of Eu^{2+} ion. However in SAED and SAEDC phosphors exhibit a

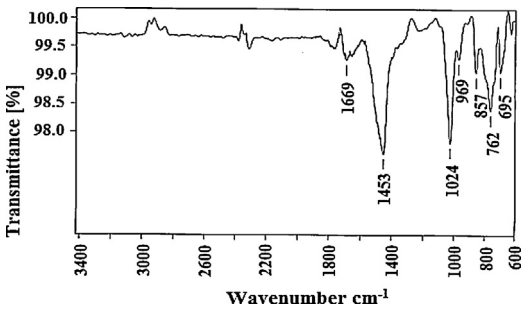

Fig. 2. FTIR spectrum of prepared $SrAl_4O_7$ phosphor.