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PREPARATION OF SR_2SIO_4: EU^{2+}, DY^{3+} NANO-PHOSPHORS PREPARED BY COMBUSTION METHOD AND ITS THERMOLUMINESCENCE PROPERTIES

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ABSTRACT

In the present paper, thermoluminescence (TL) study of Dy^{3+} doped Sr_2SiO_4:Eu^{2+} phosphor is reported. A polycrystalline sample of Sr_2SiO_4:Eu^{2+},Dy^{3+} was prepared by combustion method. The obtained phosphor was well characterized by powder X-ray diffraction, UV–visible spectroscopy and thermoluminescence . The results of the XRD show that obtained Sr_2SiO_4:Eu^{2+}, Dy^{3+} phosphor has a monoclinic structure. The average crystallite sizes could be calculated as 12.77 nm. Thermoluminescence study was carried out for the phosphor with UV irradiation here observe two glow peak .The thermoluminescence glow curves of the samples were measured at various concentration of co-dopant . With increased codopant,the intensity of the thermoluminescence peak decays and the position of the thermoluminescence peak shifts towards higher temperature, indicating the considerable retrapping associated with general order kinetics.

KEYWORDS: Thermoluminescence, Combustion Method, X-ray Emission Spectra, Nanocrystalline Materials.

1. INTRODUCTION

Rare-earth ion-doped inorganic luminescent materials have considerable applications in most devices for artificial light production (Justel et al.,1998) . Especially since the discovery of Sr_2SiO_4:Eu^{2+}, Dy^{3+} in the middle of the 1990s (Matsuzawa et al., 1996**)**. Strontium orthosilicate exits in two crystallographic modification, viz. α'-Sr_2SiO_4(orthorhombic) and β-Sr_2SiO_4(monoclinic) (Catti et al. 1983). The transition between the β-phase and the high temperature α' phase occurs at 385^0K involves the rearrangement of SiO_4 tetrahedral without disconnection of bonds (Hyde et al.1986 and Stenberg et al. 1986). With the development of scientific techniques for materials, several chemical synthesis methods, such as Sol-gel, coprecipitation, spray pyrolysis, and combustion synthesis methods, have been developed to phosphors. the combustion method has been proven to be a facile route for the low-temperature preparation of various homogenous phosphors, including silicates and aluminates, in a short time without the use of expensive high-temperature furnaces (Jia 2006, Chen et al. 1999, Jung et al. 2005 and Chakradhar et al. 2004). Thermoluminescence (TL) is an important tool for estimating the luminescent center in solids. Stimulated emission of light from an insulator or semiconductor after heating following the previous absorption of energy from ionizing radiation is known as TL (Kafadar, 2011**)**. The important application of TL materials is in TL dosimetry. In this work combustion synthesis was used to prepare Sr_2SiO_4: Eu^{2+}, Dy^{3+} phosphor using urea as fuel and NH_4Cl as flux. The properties of Sr_2SiO_4: Eu^{2+}, Dy^{3+} powders were characterized by powder X-ray diffraction (XRD), and UV–visible spectroscopy. The study of TL properties of UV rays irradiated Sr_2SiO_4:Eu^{2+} (0.01mol %), Dy^{3+} (0.01-0.05mol %) materials.

2. EXPERIMENTAL

Sr_2SiO_4: Eu^{2+}, Dy^{3+} phosphors were synthesized by combustion method. The raw materials are strontium nitrate [$Sr(NO_3)_2$ (99.99%)], silica gel (99.99%)], europium oxide[Eu_2O_3(99.99%)] and dysprosium oxide [Dy_2O_3 (99.99%)] are used as the staring materials. The starting materials are weighted according to the stoichiometry. In addition to it, Eu_2O_3 and Dy_2O_3 taken as co-activators are dissolved in concentrated nitric acid (HNO_3) before transferring them to crucible. The small amount of ammonium chloride (NH_4Cl) is used as the flux while the urea [$CO(NH_2)_2$] as a combustion fuel (Chakradhar et al. 2004 and Haranath et al., 2006). The weighed quantities of each nitrates, flux and fuel were mixed into mortar for 15 min to convert into a thick paste. The prepared paste is then placed in vertical cylindrical muffle furnace maintained at 700°C. Then the prepared samples were annealed at 800 °C for 2 h under an

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air atmosphere. The crystalline structure, size and phase composition of the sample are examined by PANanalytical using Cu-Kα radiation (λ=1.5406 Å) , where X-ray are generated at 40kV/30mA voltage and current values respectively. Absorption spectra were recorded using (Shimadzu UV-1700 UV-Visible) spectrophotometer. Thermo-luminescence was studied with PC based thermoluminescence analyzer (10091). The samples were irradiated with UV-rays source.

3. RESULT AND DISCUSSION

3.1 Optical Absorption Spectra

Optical absorption is important to study the behavior of nano-crstals and a fundamental property is the band gap. Optical excitation of electrons across the band gap is strongly allowed, producing an abrupt increase in absorption at the wavelength corresponding to the band gap energy. This feature in the optical spectrum is known as the optical absorption edge.

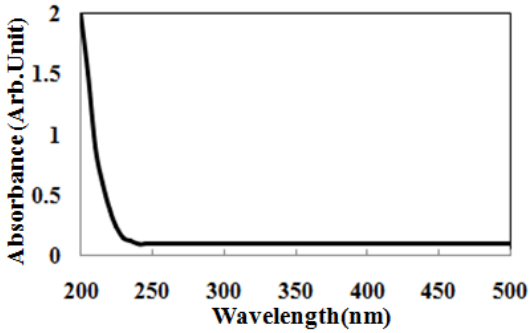

Fig 1(a) Absorption spectra of $Sr_2SiO_4:Eu^{2+},Dy^{3+}$ having absorption edge 225nm.

In figure 1(a) the optical absorption spectra of Sr_2SiO_4: Eu^{2+}, Dy^{3+} is shown in the range of 200nm-500nm.It can be seen that no absorption occurs for wavelength λ>250 nm (visible). By the absorption spectra of the sample the absorption edges was found at λ= 225 nm wavelength. The band gap obtained by plotting the graph between energy verses $(\alpha h\upsilon)^2$ shown in fig.1(b) the band gap found to be 5.5 eV.

3.2 Structural analysis : X-ray Diffraction(XRD)

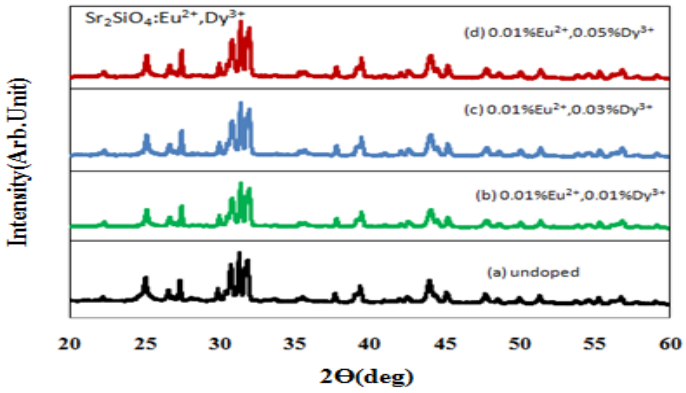

Fig.2. XRD pattern of $Sr_2SiO_4:Eu^{2+}$ phosphors with different Dy^{3+} molar concentration.

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The analysis of XRD data of Sr_2SiO_4 phase is usually qualitative, just based on relative peak intensities. It has been reported that the crystal structure of α'-Sr_2SiO_4 (orthorhombic), and β-Sr_2SiO_4 (monoclinic) are very similar (Hyde et al.1986 and Stenberg et al. 1986). The typical X-ray diffraction patterns for combustion synthesized Sr_2SiO_4: Eu^{2+} phosphor powders in presence of different Dy^{3+} concentrations are shown in figure 2 from which it can be seen that all the diffraction peaks of every samples could be indexed to the monoclinic phase of β-Sr_2SiO_4 (Ref.code 98-003-5667).

An estimation of average crystalline size for the sample is done using Scherrer's formula (Feitosa et al. 2004 and Ubale et al. 2007).

$$L = \frac{0.94\lambda}{\beta cos\theta}$$

Where L is the crystalline size, λ is the wavelength (for Cu Kα, λ=1.5406 A^0), β is the full width at half maximum (FWHM) and θ is the Bragg's angle. Table 1 shown calculation of XRD data of Sr_2SiO_4:Eu^{2+}, Dy^{3+} phosphor. The peaks in XRD patterns of different samples are similar to each other and are attributed to Sr_2SiO_4 monoclinic phase. The calculated average crystalline size of Sr_2SiO_4:Eu^{2+}, Dy^{3+} phosphor is 12.77 nm.

3.3 The Thermoluminescence Studies

Thermoluminescence is one of the possible ways to estimate the trap states of the material. Using the Chen's peak shape method (Chen, 1969), the kinetic order can be related to the geometrical factor (μ_g) by the relation $\mu_g = \delta/\omega = T_2-T_m / T_2-T_1$ where T_1, T_m and T_2 represent the temperatures of half intensity at low temperature side peak temperature and high temperature side of TL peak. And $\delta = T_2-T_m$, $\tau = T_m-T_1$ and $\omega = T_2-T_1$ Therefore E_δ , E_τ and E_ω are the corresponding activation energy.

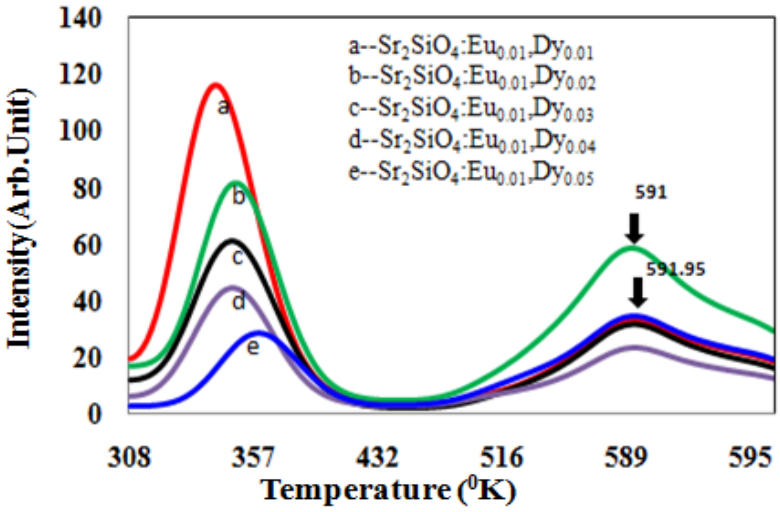

Fig. 3. TL glow curve of Sr_2SiO_4:Eu^{2+} phosphors with different Dy^{3+} concentration

Fig. 3 shows TL glow curve of Sr_2SiO_4:Eu^{2+},Dy^{3+} phosphor for different concentration of Dy, here two peak is observed due to two type of luminescence centre is formed during irradiation by UV source for 25 minutes in each

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sample. Signals increases with decreasing the doping concentration of Dy. Higher concentrations (0.05 mol%) of Dy^{3+} doped in host materials the temperature of first higher TL peak intensity is remained around 360^0K and the second small peak observe around 591^0K for other concentrations (0.02 to 0.04mol%) of Dy^{3+} doped in Sr_2SiO_4:Eu first peak and second peak remained same and is around 345^0K and 591^0K.The lowest concentration (0.01mol%) first and second peak observe around 335^0K and 591^0K .This may be due to change of phase in low concentrations of Dy^{3+}doped inSr_2SiO_4:Eu samples.

4. CONCUSION

In this work $Sr_2SiO_4:Eu^{2+}$,cDy^{3+} phosphor was prepared by combustion method which appears to be a more feasible method for production. The absorption spectra shows widened band gap because of quantum confinement effect. The quantum size effect is most pronounced nanoparticles where the band gap increases with decreasing size, resulting in the interband transition shifting to higher frequencies. XRD studies confirmed the formation of a single phase compound and find average crystalline size of $Sr_2SiO_4:Eu^{2+}$, Dy^{3+} phosphor is 12.77 nm.. The TL glow curve for Sr_2SiO_4: Eu^{2+}, Dy^{3+} expressed general order kinetics. Intensity of thermoluminescence signals decreases and peak position temperature shifts towards higher side with increasing Dy^{3+} doping concentration. The trap depth of the phosphor indicates that obey second order kinetics and this phosphor is a quite good persistent luminescent material.

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