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Research Article

Optically Stimulated Luminescence in Eu^{3+} Doped $NaLi_2PO_4$ Phosphor: Studies on Effect of Particle Size

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Abstract. We report the optically stimulated luminescence of Eu^{3+} doped $NaLi_2PO_4$ synthesized by solid state diffusion method which is crushed and sieved further using Ball milled for different time period (1 to 10 days) with different particle sizes ranging from micro to nano sizes are obtained. Formation of the material was confirmed by XRD analysis and matched peak to peak with JCPDS#80-2110. Scanning electron microscope (SEM) images were used for determining its particle size(s). Optically stimulated luminescence (OSL) characteristics were studied to see the effect of particle size after irradiating with γ-rays from Co^{60} source for wide dose range (10 Gy-100 KGy). It was found that OSL sensitivity depends on particle size of the material. Easy method of synthesis, good sensitivity, low fading, good chemical stability, etc. make this OSLD nano phosphor a good candidate for radiation dosimetry using OSL.

Keywords. $NaLi_2PO_4$, Dosimetry, Solid State Diffusion, Phosphors and Optically Stimulated Luminescence (OSL)

PACS. 87.53.Bn (Dosimetry/exposure assessment)

1. Introduction

There are lot of phosphors have been developed for high energy radiation dosimetry using Thermoluminescence (TL) and optically stimulated luminescence (OSL). There are only two OSLD phosphor are available commercially i.e. Al_2O_3:C and BeO. Due to difficult preparation method (high temperature furnace are required for carbon doping) of Al_2O_3:C make is cost effective. BeO need to take specially because its dust is very toxic and its inhaling could lung deceases called berylliosis. TL/OSL highly sensitive phosphors, are investigated as $NaMgF_3$:Eu^{2+} [5], $NaLi_2PO_4$:Eu [12], $NaLi_2PO_4$:Ce [10], SiO_2:Cu [2], $LiMgPO_4$:Tb,B [4], $Y_3Al_5O_{12}$:C [6], Na_2SiF_6:Cu,P [1], Al_2O_3:B [14], $LiAlO_2$[7], MgO:Tb [3]. However, most of them suffered with their limitations like low sensitivity, high fading, reusability issues, low dose saturation and complicated TL glow curves. Therefore, more work needed to improve dosimetric properties of existing TL/OSL phosphors and introduce some new TL/OSL. Optically stimulated luminescence (OSL) has established itself as an improved and more reliable technique for dosimetry of high-energy radiations. The OSL technique is becoming more and more popular because of several advantages over TL dosimetry, such as the completely optical nature of the instrumentation, possibility of online (in situ) measurements using optical fibre, low power consuming LED sources for stimulation, facility of re-estimation of radiation doses in case of any doubts and, overall. No structural changes due to heating that lose its reusability, unlike in some TLD phosphor materials [3, 8, 11].

In the present paper we report the synthesis of a low-Z $NaLi_2PO_4$:Eu^{3+} phosphor material. Morphology of synthesized material and Particle size effect on OSL sensitivity of $NaLi_2PO_4$:Eu^{3+} phosphor also investigated.

2. Experimental Method

$NaLi_2PO_4$:Eu^{3+} phosphor was synthesized by conventional high temperature solid state diffusion reaction. The initial materials $LiOH.2H_2O$, NaH_2PO_4 (CDH 99.5%) were taking with molar ratio 2 : 1 and the impurity salt $EuCl_3$ (also from CDH, India). The samples were prepared taking into consideration the next chemical reaction.

$$2LiOH + NaH_2PO_4 + EuCl_3 \longrightarrow Li_2NaPO_4 + NaLi_2PO_4 + H_2O$$

Firstly, LiOH and NaH_2PO_4 (in the molar ratio 2 : 1) were mixed in the presence of EtOH for better mixing in the agate mortar pestle. The mixture in powder forms was initially heated for 12 h at 673 K in quartz container in a resistive Furnace controlled by a temperature-controlled programmer (West6400) with stability around ±1 k and slowly cooled at 300 K. For final preparation of the material, it was crushed the fine particles and given heat treatment at 873 K and 1073 K for the same time interval and slowly cooled again at 300 K. The prepared sample was finally crushed in motor pestle and sieved (by using sieves of different BSS size) to obtained different particle size ranging from 25 μm-43 μm and the best particle size is obtained in between 106 μm. In the present paper, we report the systematic studied of $NaLi_2PO_4$:Eu (0.5 mol%) and synthesized by solid state diffusion method. We could prepare the whole range of particle size (ranging from micro to nano) by crushing/ball milling. The materials were irradiated by gamma radiation from ^{60}Co radioactive source for a very wide range of doses (10 Gy-100 kGy) and their OSL curves were studied. The synthesis process and Ball milling setup are shown in Figure 1.

Figure 1. Experimental route of solid state diffusion method (Ball milling)

3. Characterization

3.1 X-Ray Diffraction (XRD)

XRD pattern of the prepared $NaLi_2PO_4$:Eu^{3+} is shown in Figure 2. It could be seen from Figure 2 that the experimental data is fitted exactly with the standard data presented in JCPDF#80-2110.

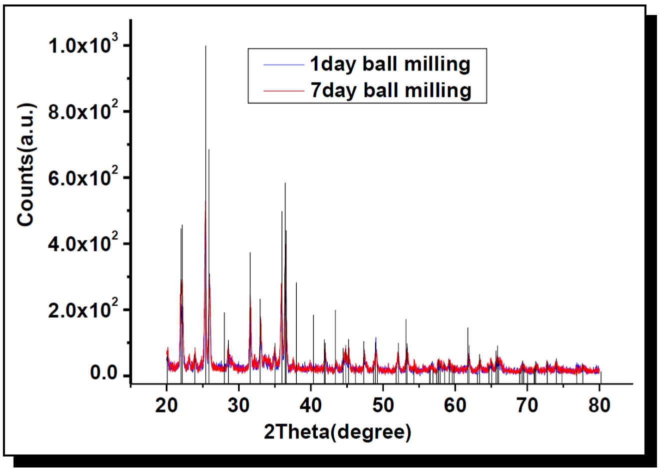

Figure 2. XRD form of the Eu doped $NaLi_2PO_4$ phosphor material. The stick pattern of the available data in the literature (JCPDS file #80-2110) is also given for comparison

No extra peak appear due to impurity and compound is seen in the figure which confirm the formation of the phosphor material. This material having direct band gap 5.169 eV. The PXRD forms were recorded on a high-resolution Bruker D8 X-ray diffractometer utilizing a monochromatic CuKα = 1.54060 Å radiation (E = 8.04 KeV) with a scan rate of 2.0/min and step size of 0.02 s. The XRD machine was operated at a voltage of 40 kV and current 25 mA. Figure 2 shows the XRD of ball milled samples of 1st day and 7th day.

3.2 Scanning Electron Microscopic (SEM) Images

SEM image shows (Figure 3) morphology of the material (0.5 mol%) having annealed at 673 K. Particle's size ranging from 250 μm-30 nm having quite uniform shapes. It could also be observed from Figure 3 that the particle size is decreasing with ball milling for longer time intervals (1 to 10 days). SEM images (Figure 3) of the material prepared through solid state diffusion method after sieves and ball milled samples were also taken. It was found that these particles are in size the ranges 250 μm-30 nm.

The ingot of $NaLi_2PO_4$:Eu prepared by high temperature conventional solid state diffusion method was crushed (using an agate motor and pestle) and sieved to get the powder samples of different particle size ranges 250 μm-30 nm. The samples in different particle size ranges were prepared by stacking the standard test sieves. Form lowers to higher pore sizes and sieving the crushed for different time intervals (from 1 to 10 days) in Teflon lined container of a ball milling machine using Zircon balls to get finer particle sizes till nano range(s). Figure 3(a-e) show SEM images of the particles ball milled for different time interval from 1 to 10 days, respectively. It images in Figure 3 show decrease in particle size with different span of ball milling. It was found from the Images 3. The average particles sizes are seen to be 250 nm, 125 nm, 80 nm, 62 nm, 30 nm for the samples ball milled foe 1, 3, 5, 7, 10 days, respectively.

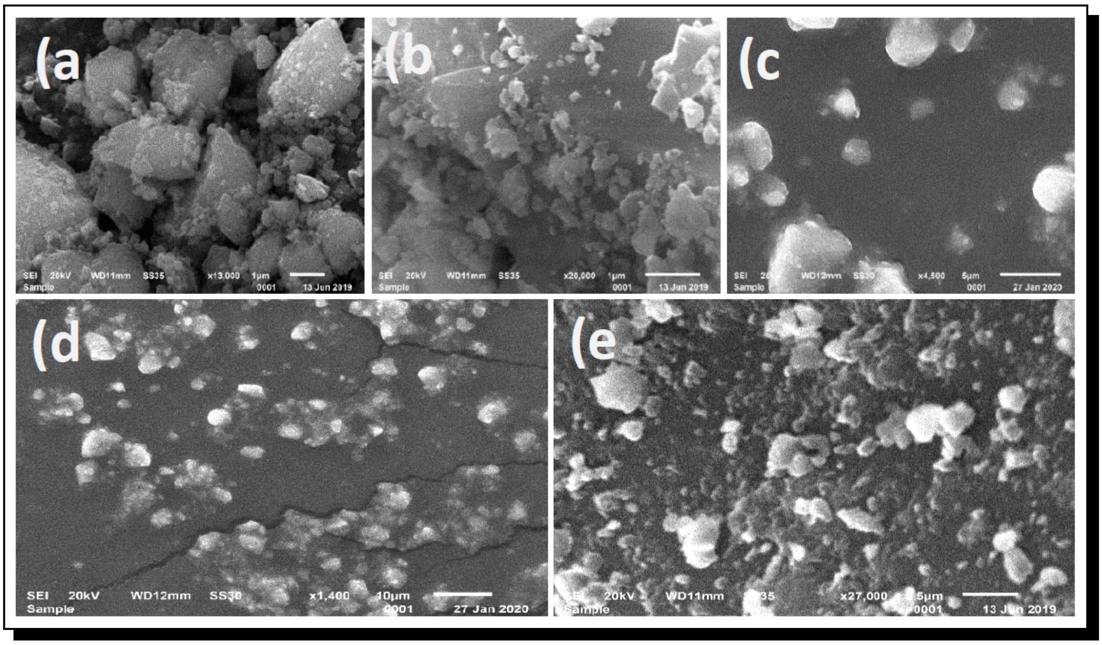

Figure 3. SEM images of SEM Images (a) 1 day, (b) 3 days, (c) 5 days, (d) 7 days, (e) 10 days ball milled samples. Indicating the decrease in particle size (from ~125 nm to ~30 nm)

3.3 Sensitivity and Dose Response

The OSL curves of Eu^{3+} doped $NaLi_2PO_4$ irradiated with γ-rays from ^{60}Co source at 10 Gy having different average particle sizes (250 μm-30 nm) and STD. Al_2O_3:C are as shown in Figure 4.

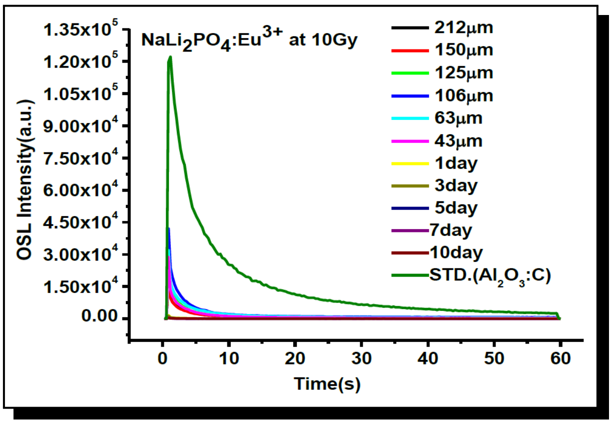

Figure 4. Typical OSL decay curves for different particle sizes of micro-to nano $NaLi_2PO_4:Eu^{3+}$ and STD. Al_2O_3:C OSLD phosphor after irradiating at 10 Gy dose of γ-rays irradiation from ^{60}Co

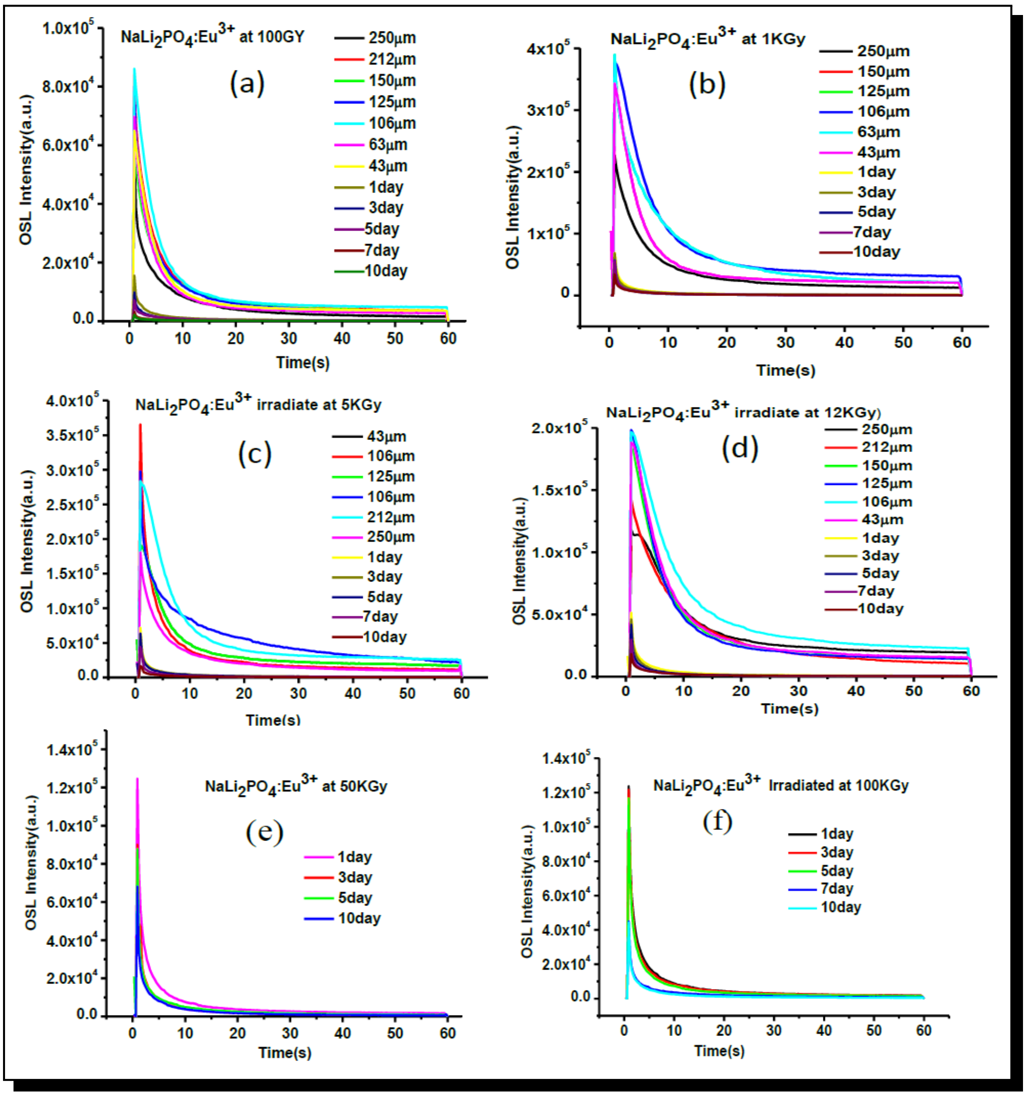

Figure 5. OSL signal variation of $NaLi_2PO_4:Eu^{3+}$ with particle size ranges (250 μm -30 nm) at different doses ranging from (10-100 kGy)

The most sensitivity of the OSLD phosphor in microcrystalline form is the particle size 63 μm-125 μm. The microcrystalline phosphor is around 3 times less sensitive than the standard

Al_2O_3:C. The nanocrystalline (30 nm) phosphor is around 106 times less sensitive than the standard Al_2O_3:C OSLD phosphor. The OSL signal variation of $NaLi_2PO_4:Eu^{3+}$ with particle size ranges (250 μm -30 nm) at different doses ranging from (10-100 kGy) are shown in Figure 5.

It can be seen from Figure 6 that for the microcrystalline particles (size range 250 μm-43 μm) the dose response is almost linear up to 1.0 kGy and start saturating thereafter. It is similar to the results already reported in our literature [13]. The main difference is that the sensitivity of the phosphor material decreases with the particle size as expected. Especially, in case of nanomaterials (average particle size range (~250 nm-30 nm). It can be seen from Figure 6 that the nanocrystalline particles dose response curve is almost linear up to 1.0 kGy and become sublinear up to 50 kGy after that it was saturating. The sensitivity is highest for the average particle size 250 nm and decreases with particle size for the lower range.

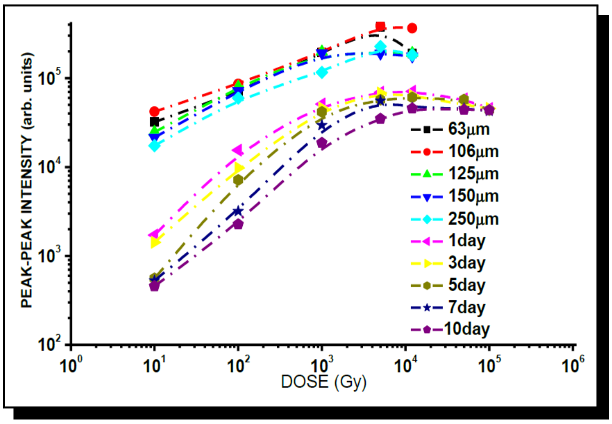

Figure 6. Dose responses (Dose vs. TL intensity (peak height) of $NaLi_2PO_4:Eu^{3+}$ phosphor material of different particle sizes irradiated at 100 to 100 kGy after irradiating with γ-rays irradiation from ^{60}Co source)

4. Fading

The fading for the macrocrystalline material 106 μm particle size has already been reported in literature [13]. Therefore, it was not studied again here. The fading of nanocrystalline at 100 Gy (~30) material was 4.8% after 20 days which is considered to be dosimetry purposes, which is even lower than that of the microcrystalline material. Thus, the nanocrystalline material also has low fading that is good for its application in radiation dosimetry using TL.

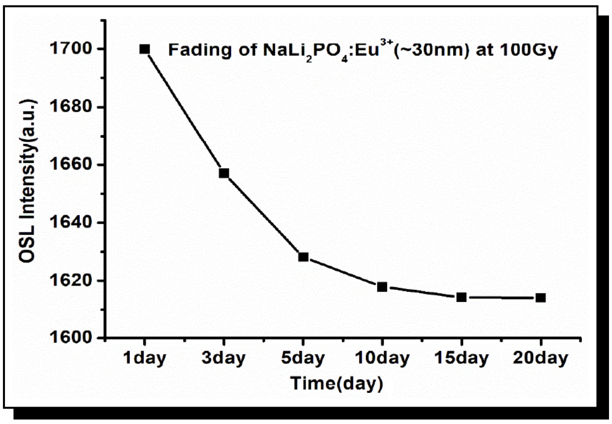

Figure 7. Fading graph of $NaLi_2PO_4:Eu^{3+}$ (~30 nm) irradiated with γ-rays irradiation from ^{60}Co source at 100Gy

5. Result and Discussion

The sample ($NaLi_2PO_4:Eu^{3+}$) was prepared by high temperature solid state diffusion. These materials ranging from micro- and nano sizes were crushing/ball milling and annealing at around 673 K for 1 h. The sensitivity of microcrystalline materials size 106 μm is 3 times less sensitive then commercially available Al_2O_3:C OSLD phosphor. On the other hand nanocrystalline materials size ~ 30nm is 106 times less sensitive than STD.Al_2O_3:C. The materials were characterized by PXRD, SEM and OSL (optically stimulated luminescence) techniques. The SEM images of ball milled samples for 1, 3, 5, 7 and 10 days having particles size 250 nm, 125 nm, 80 nm, 60 nm, 30 nm, respectively. In case of microcrystalline materials dose response liner upto 1.0 kGy thereafter decrease however, in case of nanocrystalline dose response liner upto 1.0 kGy and sublinear upto 50 kGy after that it was decrease. The fading of nanocrystalline at 100 Gy (~30) material was 4.8% after 20 days which is considered to be dosimetry purposes.

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Competing Interests

The authors declare that they have no competing interests.

Authors' Contributions

All the authors contributed significantly in writing this article. The authors read and approved the final manuscript.

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