



# Photoluminescence investigation of trivalent rare earth activated $\text{Na}_3\text{Pb}_2(\text{SO}_4)_3\text{Cl}$ phosphors for solid state lighting



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## ABSTRACT

In this paper we report the photoluminescence (PL) characteristics of  $\text{Dy}^{3+}$ ,  $\text{Sm}^{3+}$ ,  $\text{Eu}^{3+}$  and  $\text{Tb}^{3+}$  doped  $\text{Na}_3\text{Pb}_2(\text{SO}_4)_3\text{Cl}$  phosphors prepared by wet chemical method. Prepared phosphors were characterized by X-ray powder diffraction (XRD), scanning electron microscope (SEM), color co-ordinates and photoluminescence (PL) properties. The emission spectra of  $\text{Na}_3\text{Pb}_2(\text{SO}_4)_3\text{Cl}:\text{Dy}^{3+}$  phosphor show the characteristic emission of  $\text{Dy}^{3+}$  ions peaking at 478 nm and 573 nm ( $\lambda_{\text{ex}} = 390$  nm), owing to transitions of  ${}^4\text{F}_{9/2} \rightarrow {}^6\text{H}_{15/2}$  and  ${}^4\text{F}_{9/2} \rightarrow {}^6\text{H}_{13/2}$  respectively. Under 396 nm excitation the  $\text{Na}_3\text{Pb}_2(\text{SO}_4)_3\text{Cl}:\text{Eu}^{3+}$  phosphor shows emission at 594 nm and 616 nm due to  ${}^5\text{D}_0 \rightarrow {}^7\text{F}_1$  and  ${}^5\text{D}_0 \rightarrow {}^7\text{F}_2$  transitions of  $\text{Eu}^{3+}$  ions respectively. When  $\text{Na}_3\text{Pb}_2(\text{SO}_4)_3\text{Cl}:\text{Tb}^{3+}$  phosphor was excited by 380 nm, the emission spectrum showed intense green band at 545 nm due to  ${}^5\text{D}_4 \rightarrow {}^7\text{F}_5$  transition in  $\text{Tb}^{3+}$  ion. The PL emission spectrum of  $\text{Na}_3\text{Pb}_2(\text{SO}_4)_3\text{Cl}:\text{Sm}^{3+}$  phosphors by 405 nm excitation gave an emission at 562 nm ( ${}^4\text{G}_{5/2} \rightarrow {}^6\text{H}_{5/2}$ ), 598 nm ( ${}^4\text{G}_{5/2} \rightarrow {}^6\text{H}_{7/2}$ ) and 643 nm ( ${}^4\text{G}_{5/2} \rightarrow {}^6\text{H}_{9/2}$ ). The CIE color coordinates indicated that all the above phosphors were suitable as a white light-emitting phosphor. SEM studies of the phosphors show that grain size of the powders prepared by the wet chemical method is about ten micrometers range. From the obtained results it can be concluded that  $\text{Dy}^{3+}$ ,  $\text{Sm}^{3+}$ ,  $\text{Eu}^{3+}$  and  $\text{Tb}^{3+}$  doped  $\text{Na}_3\text{Pb}_2(\text{SO}_4)_3\text{Cl}$  phosphors are potential candidates for solid state lighting applications in green technology.

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## 1. Introduction

Rare-earth activated inorganic luminescence materials having special attention for developing new devices such as X-ray imaging, radiation dosimetry, solid state lasers, fiber amplifiers and biosensors [1,2]. It is well known that Nakamura et al. in 1997 developed the W-LEDs by combining blue LED chips and yellow-emitting phosphor [3]. The commercial YAG:Ce yellow phosphors became available by Nichia Chemical Co. in 1997. This phosphor when excited by blue light gives emission in yellow region [4,5]. YAG:Ce has a low color-rendering index due to lack of red component. Therefore it is necessary to develop new red phosphors having excitation in the near-UV region or blue region [6,7]. The above disadvantage could be overcome by developing a new stable phosphor by using a mixture of red and yellow emission with strong excitation band at around 400 nm, having high color rendering index. The LED light source is the best potential light sources when compared

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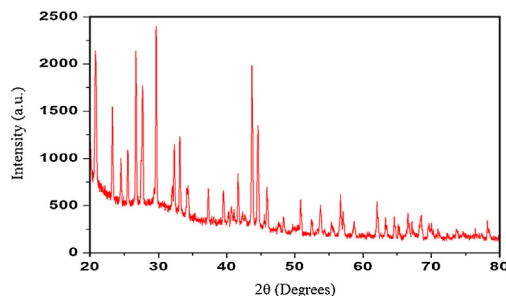


Fig. 1. XRD pattern of  $\text{Na}_3\text{Pb}_2(\text{SO}_4)_3\text{Cl}$  host.

with usual light source. LEDs have many advantages when compared with traditional light sources, such as environmental protection, an instant on–off response, relatively low cost of manufacture. They contain no mercury, have long life, space saving capacity, lower energy consumption and many other advantages [8–10]. Recently, a tremendous interest is seen in using GaN-based light-emitting diodes (LEDs) for applications in road lighting, biotechnology, interior lighting and other areas. The trivalent rare earth ions act as efficient emitters in different types of host lattices having applications in lighting industry. The currently available red phosphor  $\text{Y}_2\text{O}_3\text{:Eu}^{3+}$ , in use, shows lower efficiency that of green and blue phosphors. Most of the researchers have focused to develop the new phosphors that can be excited under the near-UV range with proper CIE coordinates and having potential application in light-emitting diodes (LEDs) [11–18]. In the recent years great efforts have been dedicated to discover many inorganic phosphors with different hosts like phosphate, silicate, aluminates and sulphate doped with different rare earth ions prepared by various synthesis methods, for solid state lighting. The trivalent rare earth ions like  $\text{Dy}^{3+}$ ,  $\text{Eu}^{3+}$ ,  $\text{Sm}^{3+}$  and  $\text{Tb}^{3+}$  were considered as the important activators for development of white light emitting diodes and lighting industry. In this paper, we have reported the  $\text{Dy}^{3+}$ ,  $\text{Sm}^{3+}$ ,  $\text{Eu}^{3+}$ ,  $\text{Tb}^{3+}$  singly doped  $\text{Na}_3\text{Pb}_2(\text{SO}_4)_3\text{Cl}$  phosphors prepared by the wet chemical method and characterized by XRD, SEM and PL characterization.

## 2. Experimental details

$\text{Na}_3\text{Pb}_2(\text{SO}_4)_3\text{Cl}$  phosphors doped with  $\text{Dy}^{3+}$ ,  $\text{Sm}^{3+}$ ,  $\text{Eu}^{3+}$  and  $\text{Tb}^{3+}$  were prepared by wet chemical method. For the preparation of  $\text{Dy}^{3+}$ ,  $\text{Sm}^{3+}$ ,  $\text{Eu}^{3+}$ ,  $\text{Tb}^{3+}$  doped  $\text{Na}_3\text{Pb}_2(\text{SO}_4)_3\text{Cl}$  phosphor, high purity  $\text{NaNO}_3$ ,  $(\text{NH}_4)_2\text{SO}_4$ ,  $\text{PbCl}_2$ ,  $\text{Tb}_4\text{O}_7$ ,  $\text{Sm}_2\text{O}_3$ ,  $\text{Dy}_2\text{O}_3$  and  $\text{Eu}_2\text{O}_3$  (99.99% purity), were used as initial raw materials. For the synthesis of  $\text{Na}_3\text{Pb}_2(\text{SO}_4)_3\text{Cl}:\text{Dy}^{3+}$ , raw materials taken in stoichiometry ratio were dissolved separately in distilled water in beaker. Then all the solutions were mixed in one beaker to obtain the desired compound. Dysprosium oxide is converted to dysprosium nitrate, by dissolving  $\text{Dy}_2\text{O}_3$  into  $\text{HNO}_3$  solution respectively. The compound  $\text{Na}_3\text{Pb}_2(\text{SO}_4)_3\text{Cl}:\text{Dy}^{3+}$ , was prepared by heating in oven at  $80^\circ\text{C}$  for 10 h and after which its powder form was obtained. The resultant white powder was crushed to fine particles in mortar pestle. Similar procedure is adopted for  $\text{Na}_3\text{Pb}_2(\text{SO}_4)_3\text{Cl}:\text{Eu}^{3+}$ ,  $\text{Na}_3\text{Pb}_2(\text{SO}_4)_3\text{Cl}:\text{Sm}^{3+}$  and  $\text{Na}_3\text{Pb}_2(\text{SO}_4)_3\text{Cl}:\text{Tb}^{3+}$  phosphors. Obtained powders were used for further study viz. XRD (X-ray diffraction), PL (Photoluminescence excitation and emission) and SEM (Scanning Electron Microscopy). The prepared host lattice was characterized for their phase purity and crystallinity by XRD using PAN-analytical diffractometer. The photoluminescence measurement of excitation and emission were recorded on the Shimadzu RF5301PC Spectrofluorophotometer. The same amount of the sample (2 gm) was used for each measurement. The emission and excitation spectra were recorded using a spectral slit width of 1.5 nm. The morphology was examined by the scanning electron microscopy (SEM, JED-2300).

## 3. Results and discussion

### 3.1. X-ray diffraction

Fig. 1, shows the X-ray Diffraction pattern of host  $\text{Na}_3\text{Pb}_2(\text{SO}_4)_3\text{Cl}$  phosphor. The obtained diffraction peaks of  $\text{Na}_3\text{Pb}_2(\text{SO}_4)_3\text{Cl}$  material do not match with any standard available data in JCPDS file. Thus it can be concluded that the obtained phase is completely new phase. This new phase is  $\text{Na}_3\text{Pb}_2(\text{SO}_4)_3\text{Cl}$  phosphor.

### 3.2. Morphology of $\text{Na}_3\text{Pb}_2(\text{SO}_4)_3\text{Cl}$ phosphor

The surface morphology of wet chemical synthesized  $\text{Na}_3\text{Pb}_2(\text{SO}_4)_3\text{Cl}$  phosphor was characterized by Scanning electron microscopy (SEM). SEM images of  $\text{Na}_3\text{Pb}_2(\text{SO}_4)_3\text{Cl}$  phosphors are shown in Fig. 2. It can be seen clearly that these morphological images are irregular in shape and size. An average particle size is in sub-micrometer range of  $\text{Na}_3\text{Pb}_2(\text{SO}_4)_3\text{Cl}$  phosphors. It is clearly observed from these SEM micrographs that the crystallite sizes are varying from one micron to ten microns.

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