

Luminescence in $\text{LaCaAl}_3\text{O}_7$ prepared by combustion synthesis

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Abstract

$\text{LaCaAl}_3\text{O}_7$ phosphors activated by several ns^2 impurities like Pb^{2+} and Bi^{3+} , and rare-earth dopants were prepared by combustion synthesis. X-ray diffraction (XRD) results confirmed the formation of single-phase compounds. Variety of activators exhibited interesting photoluminescence in $\text{LaCaAl}_3\text{O}_7$ host. Combustion synthesis furnishes a quick method for preparing these phosphors. It is suggested that $\text{LaCaAl}_3\text{O}_7$ -based materials can be developed as low-cost phosphors.

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1. Introduction

$\text{CaLnAl}_3\text{O}_7$ are complex oxides in the rare-earth calcium aluminate family with a general composition of ABC_3O_7 , where A is an alkaline earth cation, B is yttrium (Y), scandium (Sc) or a trivalent rare-earth element and C is aluminium (Al), gallium (Ga) or a transition metal ion. These compounds have melilite structure built up from CO_4 tetrahedra to form a tetragonal sheet-like arrangement; between the layers the divalent A cation and trivalent B cation are distributed randomly in eight coordinated sites with Cs symmetry. The sheet structure consists of five numbered rings of CO_4 tetrahedra perpendicular to the c -axis [1,2]. Due to the structure features, these compounds have been widely investigated as important optical materials. CaYAl_3O_7 (CYAM): Eu^{2+} , Dy^{3+} and $\text{CaGdAl}_3\text{O}_7$ (CGAM) have been studied for long-lasting phosphorescence (LLP) [3]. Kodama et al. [4,5] also studied LLP in Ce^{3+} -doped CaYAl_3O_7 . It has been reported [6] that luminescence in $\text{GdCaAl}_3\text{O}_7$: RE^{3+} ($\text{RE} = \text{Eu}, \text{Tb}$) can be excited by vacuum ultraviolet (VUV) wavelengths. This is quite important for applications like plasma display panels (PDP) and mercury-free high-intensity discharge

lamps (HID). Wang and Wang [7] included $\text{LaCaAl}_3\text{O}_7$ host in similar studies. Recently, Mahakhode et al. [8] reported X-ray-excited luminescence (XEL) in $\text{GdCaAl}_3\text{O}_7$: Eu^{3+} prepared by combustion method.

It is thus seen that there are not many studies on $\text{LaCaAl}_3\text{O}_7$ host though compounds with similar formula and structure have exhibited interesting properties. $\text{LaCaAl}_3\text{O}_7$ can be a low-cost phosphor; moreover, it may be easily prepared, like other aluminates by combustion synthesis [9,10]. It was, therefore, decided to study $\text{LaCaAl}_3\text{O}_7$ as a host. The results are reported and discussed in this paper.

2. Experimental

$\text{LaCaAl}_3\text{O}_7$ powders doped with various activators were prepared by combustion synthesis. Stoichiometric amounts of nitrates of lanthanum, calcium, aluminium and the activators were thoroughly mixed with urea. The nitrate to urea ratios were calculated by the method described earlier [9,10]. La:Ca:Al:Urea were in the ratio 1:1:3:11.66. Due to the presence of large crystallization water in aluminium nitrate, a thick paste was formed. A china dish containing the paste was inserted in a furnace heated to 500 °C in ambient air. No reducing atmosphere was provided. Within minutes the paste foamed and a flame was

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produced which lasted for several seconds. The china dish was immediately removed from the furnace. X-ray diffraction patterns were recorded on Philips PANalytical X'pert Pro diffractometer. Photoluminescence (PL) characteristics were studied using a Hitachi F-4000 spectrofluorimeter, at room temperature, using 1.5 nm spectral slit width in the range of 200–700 nm.

3. Results and discussions

Fig. 1 shows a comparison of stick patterns obtained for $\text{LaCaAl}_3\text{O}_7$ powders prepared by combustion synthesis and the corresponding pattern obtained from data file ICDD 38-1251. An excellent match is observed. $\text{LaCaAl}_3\text{O}_7$ powders are thus prepared by relatively simple combustion procedure which is initiated at 500 °C.

3.1. ns^2 activators

For ns^2 -type impurities, the ground level is 1S_0 arising from the S^2 configuration and the lowest excited levels are 3P_0 , 3P_1 and 1P_1 derived from the excited sp configuration. The absorption spectra of ns^2 ions in solids consist of three main bands labelled A, B and C in order of increasing energy corresponding to transitions $^1S_0 \rightarrow ^3P_0$, 3P_1 and 1P_1 , respectively. The C band corresponds to the allowed transition, whereas transitions corresponding to A and B bands are only partly allowed by spin–orbit coupling and vibronic coupling, respectively. The A band lies in the UV range. It is markedly sensitive to the environment [11,12]. There are very few reports available on the luminescence of ns^2 activators in aluminates [13–16] which correspond to rather old data.

Fig. 2 shows PL emission (curve a) and excitation (curve b) spectra for Pb^{2+} -doped (1 mol%) $\text{LaCaAl}_3\text{O}_7$. Emission of moderate intensity is observed in the form of a rather broad band around 372 nm. The excitation band around 268 nm is much narrower. Pb^{2+} has been studied in several hosts (for a short review see Ref. [17]). In several of these hosts the quenching temperature is quite low. Considering

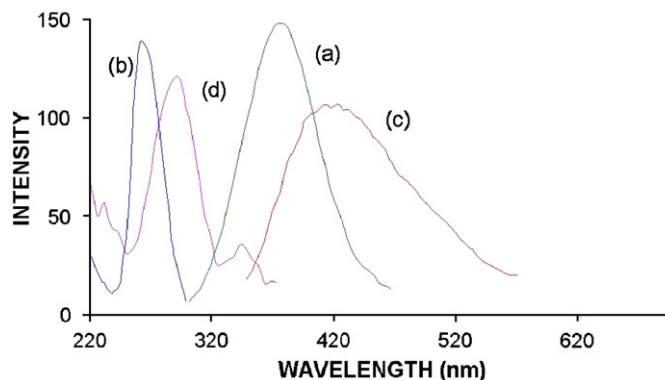


Fig. 2. Photoluminescence of Pb^{2+} and Bi^{3+} in $\text{LaCaAl}_3\text{O}_7$: (a) Pb^{2+} emission for 268 nm excitation; (b) Pb^{2+} excitation for 372 nm emission; (c) Bi^{3+} emission for 295 nm excitation; and (d) Bi^{3+} excitation for 400 nm emission.

this, the emission of even moderate intensity in $\text{LaCaAl}_3\text{O}_7$ is encouraging. The positions of the emission and excitation bands cannot be much commented upon. No systematic dependence of the position of Pb^{2+} band on the physico-chemical properties was noticed, and hence Pb^{2+} emission is said to be difficult to predict [18].

Fig. 2 also includes PL spectra for another ns^2 ion, viz. Bi^{3+} which was added in concentration of 1 mol% to $\text{LaCaAl}_3\text{O}_7$. Very broad band ranging from 350 to 600 nm and peaking around 400 nm is observed in the emission spectrum (Fig. 2, curve c). As for Pb^{2+} , the excitation band is much narrower (Fig. 2, curve d). It is located around 295 nm.

3.2. Rare-earth activators

3.2.1. Ce^{3+}

The 5d-level spectroscopy of Ce^{3+} is very simple. The 4f shell is empty and there is only one single 5d electron interacting with the crystalline environment. The Ce^{3+} ion has the $[\text{Xe}] 4f^1$ configuration, which results in only two $4f^1$ energy levels: the $^2F_{5/2}$ ground state and $^2F_{7/2}$ excited state. These energy levels are approximately 2000 cm^{-1} apart. At higher energy, the $4f^0 5d^1$ bands can be found. The energy of the bands is strongly dependent on the host lattice. The $4f^1$ ground state is separated about $51,000 \text{ cm}^{-1}$ from the excited $5d^1$ configuration. In a crystalline environment, the 5d configuration may split by as much as $25,000 \text{ cm}^{-1}$ into at most five distinct 5d states. In addition, the average energy of the five 5d levels may shift downwards by $22,000 \text{ cm}^{-1}$. The redshift of the first f–d-transition in Ce^{3+} when introduced in a crystalline host is a result of two mutually independent contributions: (1) The centroid shift, defined as the lowering of the average energy of the Ce^{3+} 5d configuration relative to the value for Ce^{3+} as a free ion. (2) The total crystal field splitting; defined as the energy difference between the lowest and highest 5d level. The $4f^0 5d^1 \rightarrow 4f^1$ emission is parity-allowed with a decay time of 3–50 ns. Both absorption and emission have a usually broad band character, showing splitting characteristic of

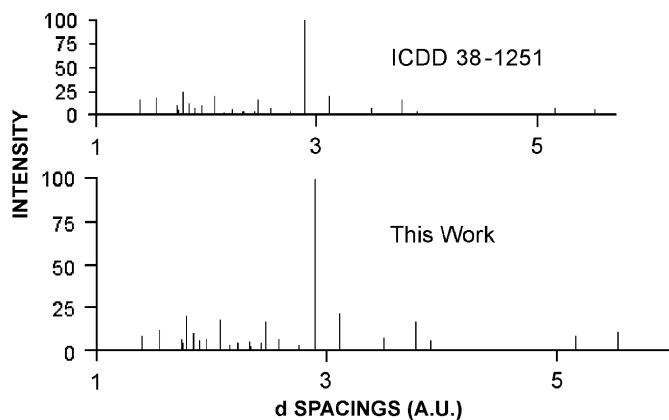


Fig. 1. Comparison of stick pattern obtained for $\text{LaCaAl}_3\text{O}_7$ with the standard data.

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