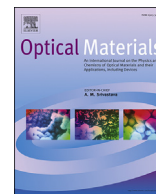




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## Optical Materials

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## Preparation and characterization of Yttrium based luminescence phosphors

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## ABSTRACTS

Ce doped Yttrium aluminate modified by replacing different molar part of aluminium or gallium (Y<sub>3</sub>Al<sub>5-x</sub>Ga<sub>x</sub>O<sub>12</sub>) and Yttrium silicate phosphors activated with Ce and Tb (Y<sub>2</sub>SiO<sub>5</sub>:Ce<sup>3+</sup>, Tb<sup>3+</sup>) were synthesized by solid state reaction and a gel combustion method, respectively. X-ray diffraction and Scanning electron microscope (SEM) techniques are used to identify their structures and morphologies. Luminescence characteristics are measured and spectroscopic data confirm that Y<sub>2</sub>SiO<sub>5</sub>:Ce<sup>3+</sup>, Tb<sup>3+</sup> phosphors can be effectively excited upon UV excitation light and X-ray irradiation, resulting in intense blue and green emissions, respectively. This energy transfer takes place by means of a non-radiative process inside Ce<sup>3+</sup>-Tb<sup>3+</sup> clusters formed in the host matrix. Tb<sup>3+</sup> doped Y<sub>2</sub>SiO<sub>5</sub> yields both blue emission <sup>5</sup>D<sub>3</sub> → <sup>7</sup>F<sub>j</sub> (j = 3,4,5,6) and green emission <sup>5</sup>D<sub>4</sub> → <sup>7</sup>F<sub>j</sub> (j = 3,4,5,6) of Tb<sup>3+</sup>. Y<sub>3</sub>Al<sub>5-x</sub>Ga<sub>x</sub>O<sub>12</sub>:Ce<sup>3+</sup> phosphors exhibit a broad blue emission band originating from allowed 5d-4f transition of the Ce<sup>3+</sup> ions under different excitation sources but the broad emission band shifts with increasing of Ga<sup>3+</sup> content. This work presents a quantitative understanding of host material's on dopant's luminescence properties and thereby provides an optimization guideline, which is extremely demanding for the development of new luminescent materials.

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## 1. Introduction

Rare-earth ion doped phosphor materials have gathered growing interests in modern life as technologically important components which are extensively applied to lighting, and plasma display panels over the past several years. Therefore, Y<sub>2</sub>SiO<sub>5</sub> has attracted much attention in recent decades due to their excellent chemical and thermal stability. When Y<sub>2</sub>SiO<sub>5</sub> having a band gap of 7.4 eV (insulator) are doped with an activator such as Ce an energy level structure occurs inside the wide band gap where the 5d to 4f transition takes place. As the shapes and positions of the emission bands are different for different activators in the host matrix the luminescence properties allow them to be an attractive luminescent material for potential applications (i.e. plasma displays [1], laser materials [2] and high energy phosphors [3]). As is well

known, Ce<sup>3+</sup> with parity allowed 4f-5d and 5d-4f transitions is not only one of the most popular activators for phosphors but also good sensitizer in co-doped materials. Ce<sup>3+</sup> shows an intense excitation and a broad yellow emission band [4–6]. Other activators such as Tb<sup>3+</sup> [7,8], Sm<sup>3+</sup> [9], Eu<sup>3+</sup> [10,11] have been widely used in the literature for this purpose. Dy<sup>3+</sup>/Tm<sup>3+</sup> [12], Eu<sup>3+</sup>/Ce<sup>3+</sup> [13], Pr<sup>3+</sup>/Ce<sup>3+</sup> [13] co-doped systems were also investigated for achieving pure white light emission. Besides, efficient energy transfer mechanisms between Tb<sup>3+</sup> and Dy<sup>3+</sup> [14], Tm<sup>3+</sup> and Dy<sup>3+</sup> [15] were reported.

Yttrium aluminium garnet (YAG), when substituted with several mole or atomic percent of the activator ion Ce<sup>3+</sup> to replace Y<sup>3+</sup>, is a luminescent material that has nearly ideal PL properties for excitation by a blue solid-state light source [16,17]. Traditional YAG:Ce phosphor is produced by conventional solid phase method at a high temperature but recently new and economical wet chemical methods such as sol-gel [18], combustion method [19] and co-precipitation [20] have been developed for synthesis of the YAG:Ce phosphor. However, the traditional solid-phase method is still dominant in all of them. A necessary condition to see the

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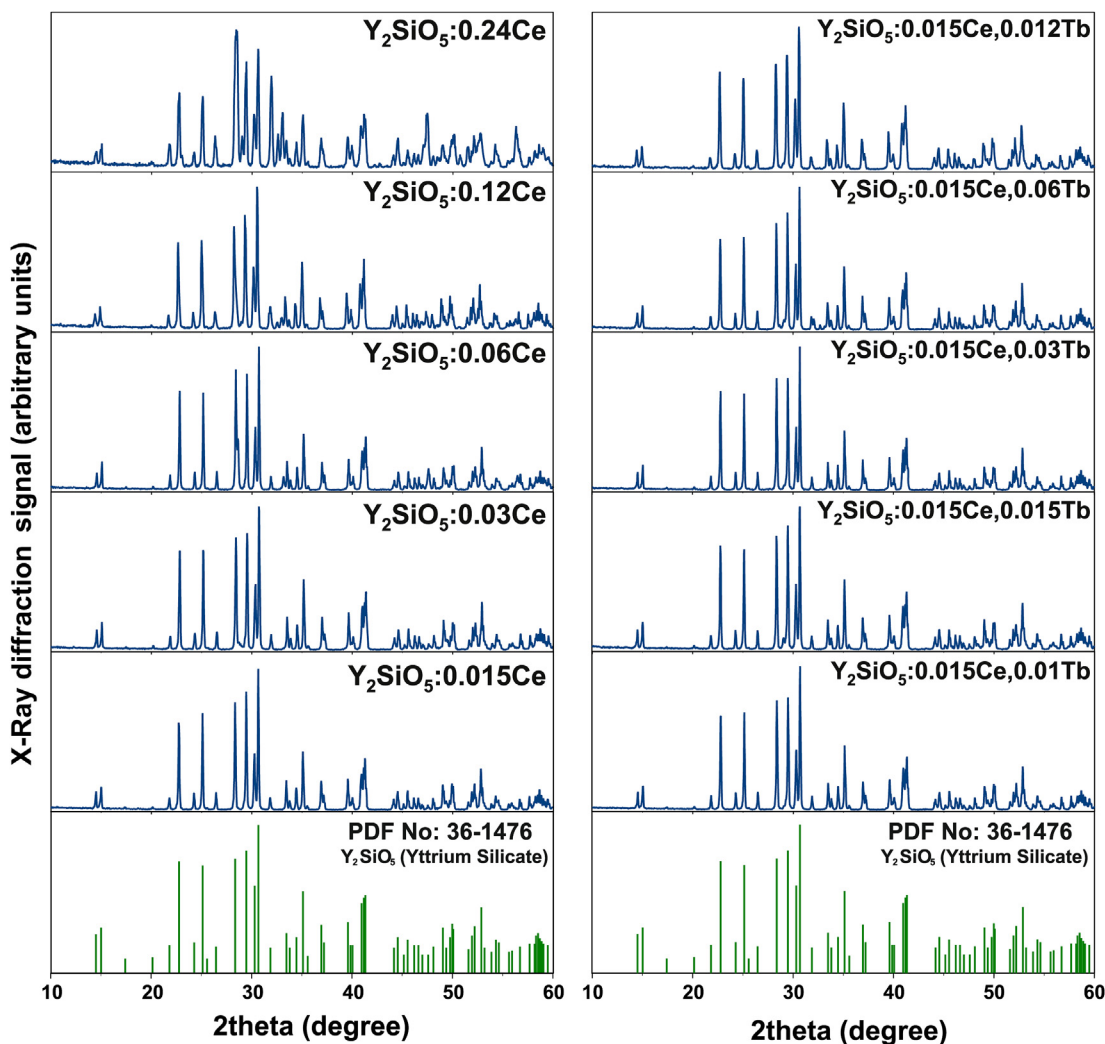


Fig. 1. XRD patterns of (a)  $\text{Y}_2\text{SiO}_5:\text{Ce}^{3+}$ ,  $\text{Tb}^{3+}$ . Reference patterns from Powder Diffraction File (PDF) are also included for comparison.

factors that govern how the YAG:Ce luminescence depends on codoping and, ultimately to be able to control the color of the YAG:Ce phosphor using codoping, is to acquire a detailed knowledge of the local structures of the double or multiple substitutional defects on the atomistic level as well as of their electronic structures [16,21,22]. From this starting point, co-dopants not only play an important role as co-activators but also as wavelength shifters [23]. For instance, substituting  $\text{Y}^{3+}$  with larger ions (e.g.  $\text{Gd}^{3+}$  and  $\text{La}^{3+}$ ) shifts the broad yellow luminescence band of  $\text{Ce}^{3+}$  to long wavelength side (red shift), which can improve color rendering properties while substitution of  $\text{Al}^{3+}$  with  $\text{Ga}^{3+}$  shifts luminescence towards a shorter wavelength (blue shift) [24,25].

This work presents studies on (i) some new understanding regarding structural and the luminescence properties of  $\text{Y}_2\text{SiO}_5:\text{Ce}$ ,  $\text{Tb}$  powders (ii) structural analysis and the luminescence characteristics of the Ce doped  $\text{Y}_3\text{Al}_{5-x}\text{Ga}_x\text{O}_{12}$  compounds excited using different excitation sources in which  $\text{Al}^{3+}$  ions were replaced with  $\text{Ga}^{3+}$  in the garnet host lattice.

## 2. Experimental

### 2.1. Materials

#### 2.1.1. Yttrium silicate ( $\text{Y}_2\text{SiO}_5$ : YSO)

Yttrium silicate phosphors single and multiple doped with

Ce, Tb, were prepared by gel combustion. Raw materials used in this study for phosphor synthesis were commercially available yttrium oxide  $\text{Y}_2\text{O}_3$  (99.9%, Aldrich), cerium nitrate  $\text{Ce}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$  (extra pure, Merck), terbium nitrate  $\text{Tb}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$  (extrapure, Merck), L-aspartic acid (99.0%, Alfa Aesar), and tetraethoxysilane (TEOS- 98% Alfa Aesar).

#### 2.1.2. Yttrium aluminate garnet ( $\text{Y}_3\text{Al}_{5-x}\text{Ga}_x\text{O}_{12}$ : YAG)

The preparation of cerium activated yttrium aluminate based phosphors was achieved by solid state reaction route from homogeneous mixtures that contain raw oxide precursors such as:  $\text{Y}_2\text{O}_3$  (Alfa Aesar 99.9%),  $\text{Al}_2\text{O}_3$  (Merk),  $\text{Ga}_2\text{O}_3$  (99.99% - Jansen);  $\text{Ce}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$  (extra pure Merck).

## 2.2. Synthesis of the phosphors

### 2.2.1. YSO: $\text{Ce}^{3+}$ , $\text{Tb}^{3+}$

The synthesis starts with the preparation of the gels from a mixture solution containing metallic nitrates, TEOS and aspartic acid. The solution mixture containing stoichiometric amounts of  $\text{Y}^{3+}$ ,  $\text{RE}^{3+}$  and  $\text{Si}^{4+}$  are prepared in order to produce phosphors with general formula  $\text{Y}_{2-x-y}\text{Ce}_x\text{Tb}_y\text{SiO}_5$ . The terbium doping concentration in terms of mole fraction is varied from 0 to 0.12. The second stage of the synthesis consists in the combustion of the gels that take place in a heating mantle at  $250^\circ\text{C}$ . The thermal synthesis of as

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