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Facile ultrasound route for the fabrication of green emitting Ba_2SiO_4: Eu^{2+} nanophosphors for display and dosimetric applications

M. Venkataravanappa[a,b], H. Nagabhushana[c,*], G.P. Darshan[d], S.C. Sharma[e,f], K.V. Archana[g], R.B. Basavaraj[c,*], B. Daruka Prasad[h]

[a] Department of Physics, Govt. First Grade College, Sira, Tumkur 572137, India
[b] Research and Development Center, Bharathiar University, Coimbatore 641046, India
[c] Prof. C.N.R. Rao Centre for Advanced Materials Research, Tumkur University, Tumkur 572103, India
[d] Department of Physics, Acharya Institute of Graduate Studies, Bangalore 560107, India
[e] Avinashilingam Institute for Home Science and Higher Education for Women University, Coimbatore 641043, India
[f] Department of Mechanical Engineering, Jain University, Bangalore 560069, India
[g] Department of Electronics and Communication Engineering, Avinashilingam Institute for Home Science and Higher Education for Women University, Coimbatore 641043, India
[h] Department of Physics, BMS Institute of Technology and Management, VTU-affiliated, Bangalore 560064, India

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ABSTRACT

A series of Eu^{2+} (1–5 mol%) activated Ba_2SiO_4 nanophosphors were prepared by ecofriendly ultrasound assisted sonochemical route using Epigallocatechin gallate (EGCG) extract as bio-surfactant. The obtained nanophosphors were well characterized by Powder X-ray diffraction (PXRD), scanning electron microscopy (SEM) etc. The average crystallite size was estimated using Debye–Scherer's formula and Williamson–Hall (W–H) plots and were found to be in the range 20–32 nm. The photoluminescence excitation spectrum exhibited two broad bands around 330 and 370 nm ascribed to the transitions of Eu^{2+} from 4f-ground state to 5d-excited state. The emission spectra showed broad band around 505 nm wavelength ascribed to 5d → 4f allowed transition of Eu^{2+} ions. Thermoluminescence (TL) of Eu^{2+} doped Ba_2SiO_4 nanophosphors were investigated using γ-irradiation in the dose range1–6 kGy at a warming rate of 5 °C min^{-1}. The phosphors show a well resolved single glow peak at ~175 °C. The TL intensity of 155 °C glow peak increase linearly with γ-dose and room temperature fading was ~76% which was highly useful in radiation dosimetry. The kinetic parameters (*E*, *s*) were estimated and the results were discussed. The chromaticity co-ordinates of all the prepared phosphors were located in green region, as a result Eu^{2+} activated Ba_2SiO_4 was a promising single phased phosphor for WLEDs.

1. Introduction

Divalent rare earth activated alkaline earth silicate phosphors are important candidates for white light-emitting-diodes (WLEDs) owing to their outstanding thermal steadiness, water and acid resistance, good thermal quenching properties and excellent luminescence properties [1–3]. Due to their exceptional properties, they are highly useful in new-generation lighting sources. WLEDs have fascinated a lot of attention due to energy saving capability, high quantum efficiencies of emission, non-toxicity, stable color, thermal and photochemical stability, extended operation lifetime and eco-friendly as a consequence they were promising candidates to substitute totally conventional incandescent and fluorescent lamps in the coming future. Currently the commercial WLED is encapsulated by the blending of a blue LED chip along with YAG: Ce^{3+} yellow phosphor [4]. Conversely, it suffers with many draw backs namely low color rendering index (CRI) and high correlated color temperature (CCT). Further, the blue chip-based white LED shows poor color stability resulting from the fluctuation of driven current [5].

The light emitted by the n-UV LEDs was generally shorter than 400 nm and show least effect on the chromatic factors of the phosphor converted LEDs, which can be investigated by means of visible (380 nm and 730 nm) radiation distribution of phosphors. Therefore n-UV phosphor converted LEDs were expected to replace conventional green LEDs generally used in the traffic signals [6–8].

* Corresponding authors. Tel:+91-9663177440
E-mail addresses: bhushanvlc@gmail.com (H. Nagabhushana), birajdar.bassu@gmail.com (R.B. Basavaraj).

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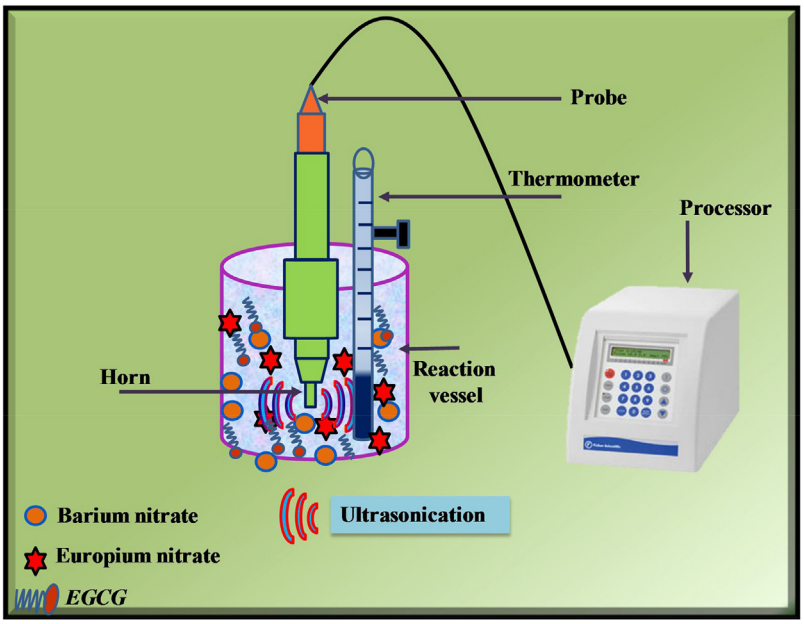

Fig. 1. Schematic representation for the synthesis of Eu^{2+} doped Ba_2SiO_4 nanophosphor by ultrasound assisted sonochemical method.

The phosphors with high absorption efficiency, high down-converting efficiency, high chemical stability and CIE color coordinates operating at different conditions mainly under forward bias-currents were paid much attention from the scientific community in the recent years [9,10].

In the present days, silicates were found to be an effective luminescent material due to their stable crystal structure, outstanding long term durability, and strong absorption in the near-UV region. Further, these silicate based phosphors exhibit better properties such as thermally stable, wide energy band gap, cost effective, non-toxic nature, and high resistance over acid, alkali and oxygen [11–17]. Traditionally, silicates were prepared at high temperature solid state method (˃1000 °C) and obtained product leads to non-uniform crystalline size and large defects which were not suitable for many applications. To overcome from these drawbacks, novel ultrasound assisted sonochemical method has been employed to synthesize silicate based nanophosphor. In ultrasound assisted sonochemical method, a series of chemical reactions arises from acoustic cavitation which results for the formation, growth and sudden collapse of bubbles in the solution. As per the hot-spot theory, a very high temperature (>5000 K) was accomplished upon the collapse of a bubble. Subsequently this collapse happens within 10^{-9} s. These exciting alterations can tune the size distribution, shape and size of the powders more effectively. In order to obtain nano/micro structured materials at sensibly low temperature was extremely indispensable for industrial applications via ultrasound route [18–22].

The allowed electric-dipole transitions between the ground state and excited state of Eu^{2+} ions were responsible for the absorption and emission spectra of Eu^{2+} doped Ba_2SiO_4 nano-phosphors [23]. The Eu^{2+} doped alkaline earth silicate based phosphors emit visible light when they absorb UV or blue light from LEDs chip. The Eu^{2+} doped phosphors were extensively investigated in the recent years due to their wide applications in white light emitting diodes (w-LEDs) [24–29].

In recent time, Eu^{2+}-doped alkaline and alkaline-earth silicate phosphors have been paid much consideration due to the lower calcination temperature and being easier to achieve electric charge balance associated with alkaline earth silicate phosphors for w-LEDs [30–33].

The bio-surfactant used in the present study was extracted from the tea leaves. In our previous paper, we have reported in detail about the extraction of EGCG extract from the tea leaves. Among the four polyphenol compounds (Epigallocatechin gallate (EGCG), Epicatechin gallate (ECG), Epigallocatechin (EGC) and Epicatechin (EC)) in the tea leaves, EGCG is the most luxuriant component in tea extract and the most potent chemically tested for biological activities [34].

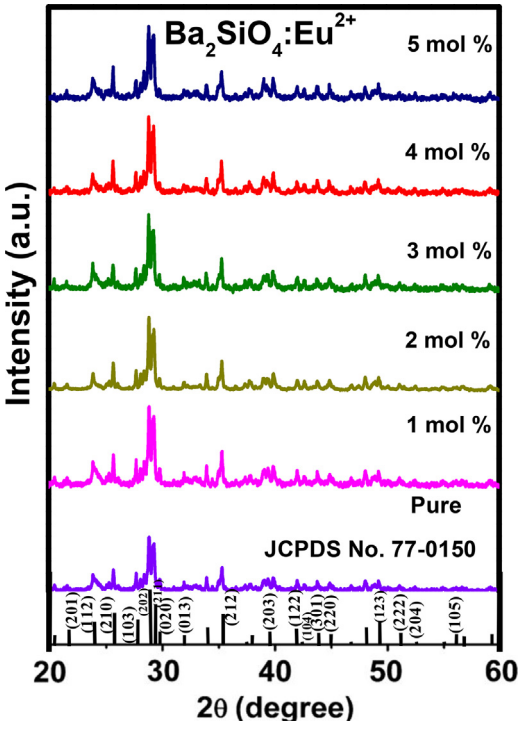

Fig. 2. PXRD patterns of pure and Ba_2SiO_4:Eu^{2+} (1–5 mol%) nanophosphor prepared with 6 h ultrasound irradiation and 30 ml EGCG extract.