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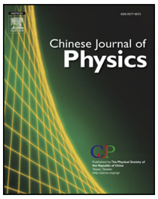

Physical, optical and radiative properties of $CaSO_4$–B_2O_3–P_2O_5 glasses doped with Sm^{3+} ions

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ABSTRACT

Trivalent rare earth ions doped borosulfophosphate glasses are in high demand owing to their several unique attributes that are advantageous for applications in diverse photonic devices. Thus, Sm^{3+} ion doped calcium sulfoborophosphate glasses with composition $25CaSO_4$–$30B_2O_3$–$(45-x)P_2O_5$–xSm_2O_3 (where $x = 0.1$, 0.3, 0.5, 0.7 and 1.0 mol%) were synthesized using melt-quenching technique. X-ray diffraction confirmed the amorphous nature of the prepared glass samples. Differential thermal analyses show transition peaks for melting temperature, glass transition and crystallization temperature. The glass stability is found in the range 91 °C to 116 °C which shows increased stability with addition of Sm_2O_3 concentration. The Fourier transform infrared spectral measurements carried out showed the presence of vibration bands due to P—O linkage, BO_3, BO_4, PO_4, P—O—P, O—P—O, S—O—B, and B—O—B unit. Glass density showed increase in value from 2.179 to 2.251 g cm^{-3} with increase in Sm_2O_3 concentration. The direct, indirect band gap and Urbach energy calculated were found to be within 4.368–4.184 eV, 3.641–3.488 eV and 0.323–0.282 eV energy ranges, respectively. The absorption spectra revealed ten prominent peaks centered at 365, 400, 471, 941, 1075, 1228, 1375, 1477, 1528 and 1597 nm corresponding to $^4D_{3/2}$,$^6H_{5/2}$→$^4I_{11/2}$,$^6P_{3/2}$, $^6F_{11/2}$, $^6F_{9/2}$, $^6F_{7/2}$, $^6F_{5/2}$, $^6F_{3/2}$, $^6H_{15/2}$ and $^6F_{1/2}$ transitions respectively. Photoluminescence spectra monitored at the excitation of 398 nm exhibits four emission bands positioned at 559, 596,643 and 709 nm corresponding to $^4G_{5/2}$→$^6H_{5/2}$, $^6H_{7/2}$, $^6H_{9/2}$ and $^6H_{11/2}$ transitions respectively. The nephelauxetic parameters calculated showed good influence on the local environment within the samarium ions site and the state of the Sm—O bond. The Judd–Ofelt intensity parameters calculated for all glass samples revealed that $\Omega_6 > \Omega_4 > \Omega_2$. The emission cross-section and the branching ratios values obtained for $^4G_{5/2}$→$^6H_{7/2}$ transition indicate its suitability for LEDs and solid-state laser application.

1. Introduction

Trivalent rare earth (RE) activated glasses are potential luminescent materials for lasers, optical detectors, sensors, light emitting diode and color displays devices applications because of their substantial emissions in the near infrared and visible region. The transitions between the 4f–4f electronic states of the rare earth ions provide emission and excitation spectra of the optical materials in the environmental field of ligand, phonon energy of the host, structure, and symmetry around the rare earth environment [1,2]. The absorption and luminescence spectra of the RE-doped glasses when related to the crystals require greater attention for technological advancement and spectroscopic measurements [3].

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More studies on the spectroscopic properties of rare earth doped glasses are still necessary for the development of efficient optical devices or to enhance the performance of the already existing ones for optoelectronic applications. This kind of investigation can provide vital information about parameters such as transition probability, the energy level structure, stimulated emission cross section and lifetime which contribute to the optical gain of the RE-doped optical devices [3].The lasing characteristics and radiative properties of the RE ions which are affected by the local perturbation around the trivalent RE ions are determined by the electronic energy states of the RE ions. For this reason, rare earth ions were doped into different network former matrices which display different laser and luminescence properties [4,5].

In recent past, researchers were focused on the rare earth doped glasses based on concentration and network former dependent spectroscopic properties via optical absorption and spectral luminescence studies to evaluate their suitability for different photonic applications. The doping of rare earth ions into glasses have numerous advantages such as hypersensitive transitions, large rare earth ion doping capability, low cost of production, large heterogeneous bandwidth, low production cost, less preparation time and easy to fabricate into different shapes when compared to crystalline materials [6]. It is necessary to find an appropriate network former for rare earth ion doping due to its vital role in the development of efficient optical devices for the fact that emission and absorption spectral intensities show considerable dependency on the ligand field environment around the rare earth ion site.

Among the glass formers, phosphate-based glasses are essential for the design of solid -state ionic and solid-state batteries because they possess unique properties such as glass transition and low melting temperature, high thermal expansion coefficient, high transparency to far infrared and UV radiations and high refractive index, etc. On the other hand, borate-based glasses are appropriate for RE^{3+} ion doping due to their essential properties such as high rare earth ion solubility, high thermal stability, and high transparency. However, pure phosphate and borate-based glasses are not stable, and their individual properties differ from the properties of combined borophosphate glasses. Phosphate-based glasses are hygroscopic in nature and with less chemical durability which affect its usefulness in the development of efficient optical devices [7,8]. Mixing B_2O_3 with P_2O_5 improves the chemical strength of the phosphate glasses by changing the glass network along with the formation of new cross-linked B—O—P bonds [9]. The borophosphate glasses have joint advantages of both the phosphate and borate glasses such as good rare earth ion solubility, good thermal and mechanical stability, excellent optical quality, narrow emission bandwidth and large emission cross section [6].

Among the rare earth ions, samarium ion is a vital activator which exhibits prominent red-orange luminescence in the visible region which is useful in visible solid-state lasers, undersea communications, color displays and high-density optical storage [10]. The Sm^{3+} ion possesses interesting properties for spectral hole burning studies and is good to analyze and understand the energy transfer between RE^{3+} and RE^{3+} ions or RE^{3+} ions and host matrix through different relaxation process [11]. The Sm^{3+} ions exhibit four dominant emission bands in the visible spectrum such as ${}^4G_{5/2}\rightarrow{}^6H_{5/2}$, ${}^6H_{7/2}$, ${}^6H_{9/2}$ and ${}^6H_{11/2}$ from its lower emitting metastable state (${}^4G_{5/2}$) with higher quantum efficiency [10]. Among the emission bands, ${}^4G_{5/2}\rightarrow{}^6H_{9/2}$ emission is associated with electric dipole allowed transition and is highly affected by the network former matrix and the strength of the transition can be changed by the ligand field around the RE-ion environment [12]. Out of the emission bands, the reddish-orange emission around 602 nm corresponding to ${}^4G_{5/2}\rightarrow{}^6H_{7/2}$ transition of the Sm^{3+} ion is useful for high-density optical storage and undersea communication applications. Furthermore, this reddish emission band is not influenced by the multi-phonon non-radiative decay since the difference in energy between the emission level and the next lower level ($\sim$7250 cm^{-1}) is considerably higher than the phonon energy of the borate glasses [13].

Swapna et al. studied the laser and spectroscopic properties of lithium fluoroborate doped Sm^{3+} glasses [10]. Aruna et al. [6], analyzed and reported the emission and absorption spectra of Sm^{3+} doped borophosphate glasses. The Sm^{3+} ions in cadmium–aluminum–silicate glasses were investigated by Yang et al. [14]. They explained the suitability for the tunable laser of the novel near-infrared emissions and near-infrared optoelectronic devices. The investigations of borosulfophosphate glasses doped Sm^{3+} ions deserve much attention due to their numerous versatility in the field of optoelectronic and laser.

This study aimed to synthesize calcium borosulfophosphate glasses doped Sm^{3+} ions and examines their structural properties by varying the rare earth ion content. The compositional roles of the $CaSO_4$–B_2O_3–P_2O_5 glasses doped with samarium ions on their optical and physical properties were assessed. Judd–Ofelt theory has been applied to calculate the radiative transition probability, Fluorescence branching ratio, and emission cross-section. Also, the photoluminescence spectra that are beneficial for fabrication of new solid-state laser devices were analyzed and compared with the existing literature.

2. Materials and method

2.1. Sample preparation

Calcium borosulfophosphate sample doped Sm^{3+} with different compositions of $25CaSO_4$–$30B_2O_3$–$(45-x)P_2O_5$–xSm_2O_3 (where x = 0.1, 0.3, 0.5, 0.7 and 1.0 in mol%) was prepared by melt quenching method. The imported pure chemicals high purity glass constituents (Sigma Aldrich 99.99%) used for this work were Calcium sulfate ($CaSO_4$), Boric acid (H_3BO_3), Phosphoric acid (H_3PO_4), and samarium oxide (Sm_2O_3) in consignments of about (30 g) were accurately weighed using standard analytical balance. The mixture of the sample was carried in an alumina crucible and then taken to an electric furnace, preheated at 200 °C for 30 min to eliminate the H_2O and H_2S content. Furthermore, the samples were heated at 1300 °C for 1 h. The melt prepared glass samples were air quenched by transferring it into a preheated stainless steel mold and kept for annealing at a rate of 300 °C for 3 h to eradicate thermal strains and then gradually allowed to cool to room temperature. After that, the glassy samples were polished to their flat surfaces for transparency and further characterization.

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