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Energy transfer from Gd^{3+} to Sm^{3+} and luminescence characteristics of $CaO–Gd_2O_3–SiO_2–B_2O_3$ scintillating glasses

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ABSTRACT

In this work, density, optical, photoluminescence and x-ray scintillation properties of calcium gadolinium silicoborate glass ($CaO:Gd_2O_3:SiO_2:B_2O_3$) have been investigated. Glasses were prepared by the conventional melt quenching technique. The results show that the density of glass increased with increasing of Sm_2O_3. The optical spectra of glass shows eleven discrete absorption bands at 360, 373, 403, 475, 944, 1077, 1225, 1368, 1468, 1520 and 1586 nm which are due to the transitions of $^6H_{5/2}$ to $^4D_{3/2}$, $^6P_{7/2}$, $^6P_{3/2}$, $^4I_{11/2}$, $^6F_{11/2}$, $^6F_{9/2}$, $^6F_{7/2}$, $^6F_{5/2}$, $^6F_{3/2}$, $^6H_{15/2}$ and $^6F_{1/2}$, respectively. The emission spectra were observed and assigned to 312, 563, 600, 646 and 703 nm by excitation at 275 nm. The emission intensity of Sm^{3+} increased with increasing of Sm_2O_3 concentration until 0.25 mol%, while decay time decrease with increasing of Sm_2O_3 content. For results of the radioluminescence (RL), they perform four emission peaks with the strongest emission at 598 nm. Glass doped with 0.35 mol% Sm_2O_3 show the highest RL emission. The integral scintillation efficiency of 0.35 mol% Sm_2O_3 doped glass was determined as 25% of commercial BGO scintillator crystal.

1. Introduction

Glass scintillator can be used for detection of X-rays and neutrons. Glass material performs the variety of advantage such as easy synthesis, low-price, high optical homogeneity, large volume production and various shaping [1,2]. Borate glasses are the technologically important glass formers and play a significant role in various applications. Furthermore, increase of B_2O_3 content from 10 mol% to 30 mol% resulted in the emergence of $[BO_3]$ units and Non-Bridging Oxygen (NBOs) growth [3–5]. In part of the silicate glasses, they were used as host material for luminescence study of rare-earth and transition metal ions, due to they have good optical and mechanical properties as well as good chemical durability [6].Cooperative of these two glass formers, borosilicate glasses perform low melting temperature, low coefficients of thermal expansion and can be used in relatively high temperatures [7]. The effect of adding calcium, it can increase intensity of optical absorption and luminescence emission in glass [8]. For rare earth oxides, intensive Gd_2O_3 are popular material due to the efficient energy transfer from Gd^{3+} ions to the luminescence activators, high thermal neutron capture cross-section and increase the light yield of emission [9,10]. In recent years, glass scintillators containing high Gd_2O_3 contents have been succeed in various germinate, phosphate, silicate and borosilicate glasses with fast decay time [1,11]. Glasses containing Sm^{3+} ions have the extensive interest due to their potential application for high-density optical storage, under sea communication and color displays [12]. The optical properties of Sm^{3+} doped glasses have attracted much attention because of their technological applications. Additionally, the Sm^{3+} ions exhibit broad emission bands due to $^4G_{5/2} \rightarrow {}^6H_J$ ($J=5/2$, 7/2, 9/2, 11/2) transitions in any host matrix [13]. It is also well known that the intensities of emission bands of Sm^{3+} ion in glasses depend on its concentration and glass composition [14]. Several literatures were published luminescence properties of Sm^{3+} in different glass host such as tellurite, phosphate, silicate and borate glasses [13,15–20].

In this work, a new series of $CaO:Gd_2O_3:SiO_2:B_2O_3$ glass have been prepared by melted-quenching technique. Sm_2O_3 was doped in $CaO:Gd_2O_3:SiO_2:B_2O_3$ glasses with different concentrations for study in physical, optical, luminescence properties and scintillation efficiency. This glass has never been studied in any detail and

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this work is the first time for understanding the effects of glass composition and Sm_2O_3 concentration on properties in CaO:Gd_2O_3:SiO_2:B_2O_3 glass system, developed for using to glass scintillator.

2. Experiment

Glasses compositions of 10CaO:25Gd_2O_3:10SiO_2:(55-x)B_2O_3:$x$$Sm_2O_3$ (where x are 0.05, 0.15, 0.25, 0.35, 0.45, 0.50 and 1.00 mol%) were prepared by melt quenching technique. The high purity chemicals, CaO, Gd_2O_3, SiO_2, H_3BO_3 and Sm_2O_3 were mixed thoroughly in an alumina crucible. Each batch of formulas was weighted to 30 g melted in an electric furnace at a temperature of 1400 °C for 3 hours. The melted material was quenched in pre-heated stainless-steel molds, which would yielded several batches of glass samples. The glass samples were further annealed at 550 °C for 3 hours in order to remove thermal strains. Glasses were cut and polished to $1.0 \times 1.5 \times 0.3$ cm^3 for property investigations. The density measurement was applied by Archimedes's principle, glasses were weighted in air and water as an immersion liquid using a 4-digit sensitive microbalance (AND, HR-200). The optical spectra of glass samples were measured with a UV–vis–NIR spectrophotometer (Shimadzu UV-3600) in the wavelength range 350–1800 nm. The excitation, emission spectra and decay curves were recorded by using a spectrofluorophotometer (Cary-Eclipse) with xenon lamp as a light source. The non-exponential decay curves were fitted, to evaluate energy transfer parameter (Q), with the Inokuti–Hirayama (IH) model

$$I(t) = I_0 exp\left\{-\frac{t}{\tau_0} - Q\left(\frac{t}{\tau_0}\right)^{3/s}\right\}$$

where $I(t)$ is fluorescence intensity at time after excitation (t), I_0 is fluorescence intensity at initiate time, and τ_0 is the intrinsic decay time of the donors in the absence of acceptors. The value of S (6, 8 or 10) depends on whether the dominant mechanism of the interaction is dipole–dipole, dipole–quadrupole or quadrupole–quadrupole, respectively [21,22]. The X-ray luminescence spectra of glasses were measured with a Cu target X-ray generator (Inel, XRG3D-E), whose X-ray source was operated at 50 kV and 30 mA, and the spectrometer (QE65 Pro, Ocean Optics) with an optical fiber to detect the emission spectra, as shown in Fig. 1.

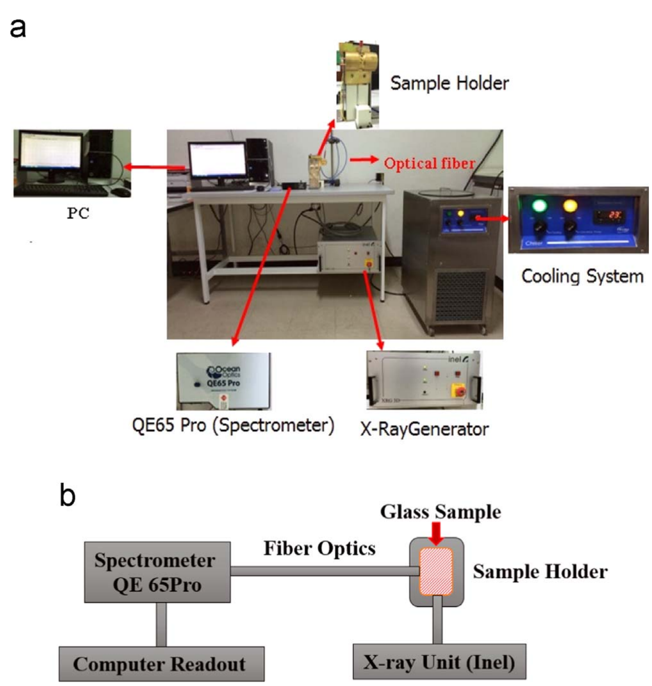

Fig. 1. (a) Experimental setup and (b) schematic diagram of the X-ray induced optical luminescence spectrometer.

3. Results

All the Sm^{3+} doped CaO:Gd_2O_3:SiO_2:B_2O_3 glasses exhibit soft yellow color and high transparency, as shown in Fig. 2.

4. Density

Glass density tend to increase with increasing of Sm_2O_3 concentration, as shown in Fig. 3. It means that there is a change in the structural arrangement of the atoms with Sm_2O_3 addition in CaO:Gd_2O_3:SiO_2:B_2O_3 network which shows that borate is replaced by samarium oxide since the density of Sm_2O_3 is 8.347 g/cm^3 while that of B_2O_3 is 2.460 g/cm^3. The increased density of the samples is due to higher molecular weight of samarium than any other component of the given glass system.

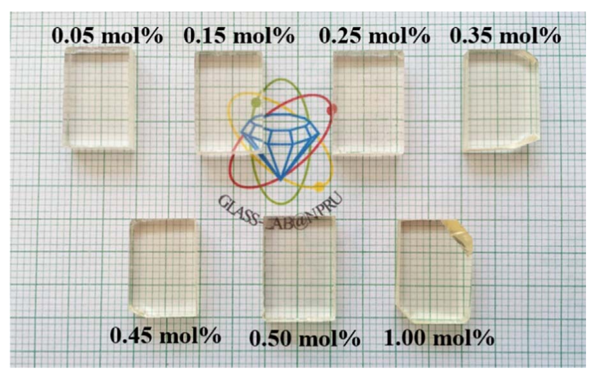

Fig. 2. The Sm^{3+} doped CaO:Gd_2O_3:SiO_2:B_2O_3 glasses. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

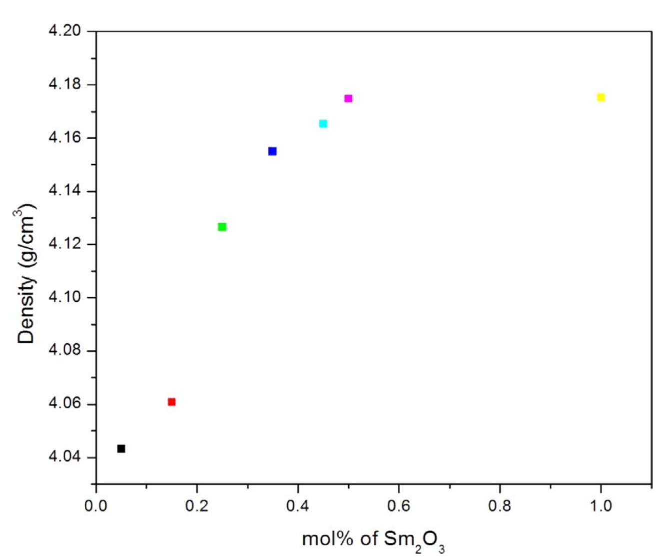

Fig. 3. The density of the Sm_2O_3 doped CaO:Gd_2O_3:SiO_2:B_2O_3 glasses.